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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Inventor	: Andrew J. Callinan	Appeal No.
Appln. No.	: 10/725,634	
Filed	: December 2, 2003	Group Art Unit: 1771
Title	: CATIONIC MICROSPHERES AND METHOD OF MAKING CATIONIC MICROSPHERES	Examiner: Daniel R. Zirker
Docket No.	: 58876US002	

RESPONSE TO NOTICE OF NON-COMPLIANT APPEAL BRIEF

Mail Stop Appeal Brief - Patents
Commissioner For Patents
P.O. Box 1450
Alexandria, VA 22313-1450

SENT VIA EXPRESS MAIL
Express Mail No.: EQ 141629629 US

Sir:

This Response is submitted in reply to the Notice Of Non-Compliant Appeal Brief mailed on October 13, 2006. In the Notice Of Non-Compliant Appeal Brief, the Examiner checked the Box 5, which states: "The brief does not contain a concise statement of each ground of rejection presented for review (37 CFR 41.37(c)(1)(vi))." The Examiner also checked Box 10 on the Notice Of Non-Compliant Appeal Brief and provided the following further additional argument:

More particularly, in connection with Box 5, appellants have on page 8 of the Brief in the Summary section improperly characterized the Examiner's rejection of claims 1, 4, 7-10, 29, 36, and 37 in view of Howard as being separately anticipated and also obvious, whereas in point of fact the Examiner has rejected these claims as being either anticipated or obvious in view of the reference.

Despite the Examiner's comments, Applicants do not believe the separate statement of the anticipation and obviousness rejection based on U.S. Patent No. 4,598,112 to Howard constitutes a mischaracterization of the Examiner's rejection. Rather, the separate statement of the two components of the rejection was made for purposes of organizing the Appeal Brief, and not for purposes of characterizing the Examiner's rejection as an anticipation rejection and an obviousness rejection. Applicants believe it is evident from the Appeal Brief, particularly since Applicants recited the Examiners rejection in the

alternative at the top of page nine of the Appeal Brief, that Applicants approach to organizing the brief was not misleading as to the alternative nature of the Examiner's rejection.

Nonetheless, as requested by the Examiner, Applicants have renumbered former Issues I and II as Issue I with subparts (a) and (b) on page 8 of the attached Amended Appeal Brief and elsewhere in the Amended Appeal Brief and have consequently renumbered former Issue III as Issue II on page 8 of the attached Amended Appeal Brief and elsewhere in the Amended Appeal Brief. These changes are believed to further emphasize the alternative nature of the Examiner's rejection based on the Howard patent. Furthermore, these changes are believed to fully and adequately address the Examiner's contentions at Boxes 5 and 10 of the Notice of Non-Compliant Appeal Brief.

Consideration and entry of the attached Amended Appeal Brief is respectfully requested. Furthermore, reconsideration and reversal of the rejections of claims 1-51 accompanied by allowance of claims 1-51 are respectfully requested.

Respectfully submitted,

Date: January 8, 2007

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Title : CATIONIC MICROSPHERES AND METHOD
OF MAKING CATIONIC MICROSPHERES

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Examiner: Daniel R. Zirker

AMENDED BRIEF FOR APPELLANT

Mail Stop Appeal Brief - Patents
Commissioner For Patents
P.O. Box 1450
Alexandria, VA 22313-1450

SENT VIA EXPRESS MAIL
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Sir:

This is an appeal from an Office Action dated January 30, 2006 in which the Examiner finally rejected all of the claims present in the above-identified application, namely claims 1-51.

REAL PARTY IN INTEREST

The real party in interest in this Appeal is 3M Innovative Properties Company, which is the owner of the entire right, title, and interest in the above-identified application.

RELATED APPEALS AND INTERFERENCES

There are no known related appeals or interferences involving the subject matter or issues in this Appeal, and there are no known related appeals or interferences that will directly affect, be directly affected by, or have a bearing on the Board's decision in this Appeal.

STATUS OF THE CLAIMS

A. TOTAL NUMBER OF CLAIMS IN THE APPLICATION

Claims in the application are: Claims 1-51.

B. STATUS OF ALL THE CLAIMS

1. Claims canceled:
None.
2. Claims withdrawn from consideration, but not canceled:
None.
3. Claims pending:
Claims 1-51.
4. Claims allowed:
None.
5. Claims rejected:
Claims 1-51.

C. CLAIMS ON APPEAL

1. The claims on appeal are: Claims 1-51.

STATUS OF AMENDMENTS

No amendments were submitted after the Final Rejection contained in the Office Action dated January 30, 2006.

SUMMARY OF INVENTION

Independent claim 1 defines an adhesive composition. The adhesive composition of claim 1 comprises a plurality of cationic microspheres and a fluidizing medium. According to claim 1, the fluidizing medium is effective for supporting fluid application of the adhesive composition to a surface. See lines 28-30 on page 2, lines 13-17 on page 3, lines 18-26 on page 18, and original claim 1.

Claim 2 depends from independent claim 1 and specifies the cationic microspheres have a cationic charge that is permanently and individually bound to each cationic microsphere. See lines 18-26 on page 18 and original claim 2.

Claim 3 depends from independent claim 1 and specifies the cationic microspheres are derived in part from a cationic unsaturated vinyl monomer. See lines 6-10 on page 3, line 21 on page 4 through line 8 on page 5, and original claim 3.

Claim 5 depends from independent claim 1 and specifies the cationic microspheres are derived in part from an unsaturated vinyl comonomer. See lines 10-12 on page 3, line 9 on page 4 through line 28 on page 7, and original claim 5.

Claim 6 depends from independent claim 1 and specifies the adhesive composition further comprises a cationic latex adhesive binder. See lines 29-31 on page 3, line 7 on page 11 through line 6 on page 12, and original claim 6.

Claim 7 depends from independent claim 1 and specifies the adhesive composition further comprises a cationic surfactant. See lines 14-17 on page 3, lines 14-32 on page 8, and original claim 7.

Independent claim 11 defines a mixture that comprises polymerizable substances. The polymerizable substances of claim 11 comprise at least one C_N alkyl(meth)acrylate monomer, where N is any integer ranging from 4 to 14, and a cationic unsaturated vinyl comonomer. See lines 6-10 and lines 13-17 on page 3 and original claim 11.

Claim 13 depends from independent claim 11 and specifies the polymerizable substances further comprise an unsaturated vinyl comonomer. See lines 10-12 on page 3, line 9 on page 4 through line 28 on page 7, and original claim 13.

Claim 14 depends from independent claim 11 and defines a coating composition. The coating composition of claim 11 comprises cationic microspheres based on the mixture of independent claim 11 and a cationic latex adhesive binder. See lines 27-31 on page 3, line 7 on page 11 through line 6 on page 12, and original claim 14.

Claim 15 depends from dependent claim 14 and further defines the coating composition. According to claim 15, the mixture further comprises a cationic surfactant. See lines 14-17 on page 3, lines 14-32 on page 8, original claim 15, and claim 15, as amended via the Amendment filed on December 1, 2005.

Claim 16 depends from independent claim 11 and specifies the mixture further comprises a cationic surfactant. See lines 14-17 on page 3, lines 14-32 on page 8, and original claim 16.

Claim 18 depends from independent claim 11 and specifies that polymerization of the polymerizable substances is effective to form cationic microspheres. See lines 13-17 and lines 23-27 on page 3, lines 18-26 on page 18, original claim 18, and claim 18, as amended via the Amendment filed on December 1, 2005.

Independent claim 19 defines an adhesive that comprises a polymerized product of polymerizable substances. Claim 19 states the polymerized product is adhesively repositionable between different application surfaces. According to claim 19, the polymerizable substances comprise at least one C_N alkyl(meth)acrylate monomer, where N is any integer ranging from 4 to 14; and a cationic unsaturated vinyl comonomer. See lines 28-30 on page 2, lines 13-17 on page 3, lines 18-26 on page 18, and original claim 19.

Claim 22 depends from independent claim 19 and specifies the polymerizable substances further comprise an unsaturated vinyl comonomer. See lines 10-12 on page 3, line 9 on page 4 through line 28 on page 7, and original claim 22.

Claim 23 depends from independent claim 19 and specifies the adhesive further comprises a cationic latex adhesive binder. See lines 29-31 on page 3, line 7 on page 11 through line 6 on page 12, and original claim 23.

Claim 24 depends from independent claim 19 and specifies the adhesive further comprises a cationic surfactant. See lines 14-17 on page 3, lines 14-32 on page 8, and original claim 24.

Claim 25 depends from independent claim 19 and specifies performance capabilities of the adhesive. According to claim 25, the adhesive is coated as an adhesive stripe about 18 mm wide and about 33 mm long on a paper substrate at the rate of about 0.65 grams of the adhesive per square foot (7.0 grams per square meter) of the adhesive stripe. Claim 25 states the coated adhesive, when subjected to static angle testing on a surface of a painted steel panel, detaches from the surface of the painted steel panel about 300 seconds after being adhered to the surface of the painted steel panel. See line 19 on page 20 through line 19 on page 21, Example 1 (line 27 on page 21 through line 25 on page 23), line 29 on page 25 through line 17 on page 27 (see Table 1 on page 27), and original claim 25.

Claim 26 depends from claim 23 and further specifies performance capabilities of the adhesive. According to claim 26, the adhesive is coated as an adhesive stripe about 18 mm wide and about 33 mm long on a paper substrate at the rate of about 0.65 grams of the adhesive per square foot (5.9 grams per square meter) of the adhesive stripe. Claim 26 states the coated adhesive, when subjected to static angle testing on a surface of a painted steel panel, detaches from the surface of the painted steel panel about 240 seconds after being adhered to the surface of the painted steel panel. See line 19 on page 20 through line 19 on page 21, Example 2 (line 27 on page 23 through line 27 on page 25), line 29 on page 25 through line 17 on page 27 (see Table 1 on page 27), and original claim 26.

Independent claim 29 defines a method of an adhesive composition. The method of claim 29 comprises causing a plurality of cationic microspheres to exist in a fluidizing medium, which yields the adhesive composition. According to claim 29, the fluidizing medium is effective for supporting fluid application of the adhesive composition to a surface. See lines 28-30 on page 2, lines 13-17 on page 3, lines 18-26 on page 18, and original claim 29.

Claim 30 depends from independent claim 29 and specifies the cationic microspheres have a cationic charge that is permanently and individually bound to each cationic microsphere. See lines 18-26 on page 18 and original claim 30.

Claim 32 depends from independent claim 29 and specifies the cationic microspheres are derived in part from an unsaturated vinyl comonomer. See lines 10-12 on page 3, line 9 on page 4 through line 28 on page 7, lines 17-22 on page 12, and original claim 32.

Claim 33 depends from independent claim 29 and further specifies incorporating a cationic latex adhesive binder in the adhesive composition. See lines 29-31 on page 3, line 7 on page 11 through line 6 on page 12, lines 5-9 on page 15, and original claim 33.

Claim 34 depends from claim 33 and further specifies incorporating a cationic surfactant in the adhesive composition of claim 33. See lines 14-17 on page 3, lines 14-32 on page 8, lines 17-25 on page 12, and original claim 34.

Claim 35 depends from independent claim 29 and specifies incorporating a cationic surfactant in the adhesive composition of claim 29. See lines 14-17 on page 3, lines 14-32 on page 8, lines 17-25 on page 12, and original claim 35.

Independent claim 38 defines a method of making adhesive cationic microspheres. According to the method of claim 38, a polymerizable mixture comprises at least one C_N alkyl(meth)acrylate monomer, where N is any integer ranging from 4 to 14, and a cationic unsaturated vinyl comonomer. The adhesive cationic microspheres are made in accordance with the method of claim 38 by initiating reaction of the polymerizable mixture. See lines 6-10, lines 13-17, and lines 23-24 on page 3 and original claim 38.

Claim 40 depends from independent claim 38 and specifies the polymerizable mixture further comprises an unsaturated vinyl comonomer. See lines 10-12 on page 3, line 9 on page 4 through line 28 on page 7, lines 17-22 on page 12, and original claim 40.

Claim 43 depends from independent claim 38 and specifies the method of making adhesive cationic microspheres further comprises including a cationic surfactant in the polymerizable mixture. See lines 14-17 on page 3, lines 14-32 on page 8, lines 17-25 on page 12, and original claim 43.

Claim 44 defines a method of using the adhesive cationic microspheres of claim 38. According to claim 44, the method comprises uniformly mixing the adhesive cationic microspheres with a cationic latex adhesive binder to form an adhesive composition. See lines 27-31 on page 3, line 7 on page 11 through line 6 on page 12, lines 5-9 on page 15, and original claim 44.

Claim 45 depends from claim 44 and further specifies including a cationic surfactant in the polymerizable mixture. See lines 14-17 on page 3, lines 14-32 on page 8, lines 17-25 on page 12, and original claim 45.

Claim 46 depends from claim 45 and specifies performance capabilities of the adhesive composition. According to claim 46, the adhesive composition is coated as an adhesive stripe about 18 mm wide and about 33 mm long on a paper substrate at the rate of about 0.65 grams of the adhesive composition per square foot (7.0 grams per square meter) of the adhesive stripe. Claim 46 states the coated adhesive composition, when subjected to static angle testing on a surface of a painted steel panel, detaches from the surface of the painted steel panel about 300 seconds after being adhered to the surface of the painted steel panel. See line 19 on page 20 through line 19 on page 21, Example 1 (line 27 on page 21 through line 25 on page 23), line 29 on page 25 through line 17 on page 27 (see Table 1 on page 27), and original claim 46.

Claim 47 defines a method of using the adhesive cationic microspheres of claim 40. According to claim 47, the method comprises uniformly mixing the adhesive cationic microspheres with a cationic latex adhesive binder to form an adhesive composition. See lines 27-31 on page 3, line 7 on page 11 through line 6 on page 12, lines 5-9 on page 15, and original claim 47.

Claim 48 depends from claim 47 and further specifies including a cationic surfactant in the polymerizable mixture. See lines 14-17 on page 3, lines 14-32 on page 8, lines 17-25 on page 12, and original claim 48.

ISSUES

- I. One issue is whether claims 1, 4, 7-10, 29, 36 and 37 are either:
- (a) anticipated under 35 U.S.C. §102(b) by U.S. Patent No. 4,598,112 to Howard (“the Howard patent”) or
 - (b) rendered obvious under 35 U.S.C. 103(a) by the Howard patent.
- II. Another issue is whether claims 1-51 are rendered obvious under 35 U.S.C. 103(a) by U.S. Patent No. 4,598,112 to Howard (“the Howard patent”) in combination with U.S. Patent No. 5,756,625 to Crandall et. al. (“the Crandall patent”).

ARGUMENT

I. (a). The Howard Patent Does Not Anticipate Claims 1, 4, 7-10, 29, 36 and 37.

A. The Examiner failed to establish that the Howard patent discloses each and every element of claims 1 and 29.

The Examiner has alleged the Howard patent anticipates claims 1 and 29 under 35 U.S.C. §102(b). However, despite the Examiner’s statements in support thereof, the Howard patent does not in fact anticipate any of claims 1 or 29 under 35 U.S.C. §102(b).

"For a prior art reference to anticipate in terms of 35 U.S.C. §102, every element of the claimed invention must be identically shown in a single reference." In re Bond, 15 U.S.P.Q.2d 1566, 1567 (Fed. Cir. 1990). Furthermore, the elements disclosed by the single reference "must be arranged as in the claim under review." Bond, 15 U.S.P.Q.2d at 1567. Moreover, "[i]n deciding the issue of anticipation, the trier of fact must identify the elements of the claims . . . and identify corresponding elements disclosed in the allegedly anticipating reference." Lindemann Maschinenfabrik GMBH v. American Hoist and Derrick, 221 U.S.P.Q. 481, 485 (Fed. Cir. 1984). Thus, "it is incumbent upon the Examiner to identify wherein each and every facet of the claimed invention is disclosed in the applied reference." Ex parte Levy, 17 U.S.P.Q.2d 1461, 1462 (P.T.O. Bd. Pat. App. & Int’f 1990).

In support of this rejection, the Examiner initially provided the following comments:

Claims 1, 4, 7-10, 29, 36 and 37 are rejected under 35 U.S.C. 102(b) as anticipated by or, in the alternative, under 35 U.S.C. 103(a) as obvious over Howard. Note particularly the Abstract, Col 1, line 28- Col 2, line 15, Claim 1. The reference discloses a repositionable acrylate adhesive composition, coated articles and methods of making the adhesive wherein the adhesive may simply be a plurality of cationic microspheres immersed in a film forming binder, which is substantially all that at least the majority of these claims require. As to claims 9 and 37, the absence of adhesive residue is believed to be either inherent, or alternatively an obvious modification to one of ordinary skill.

First complete paragraph on page 3 of Office Action dated September 1, 2005. Despite these comments, the Howard patent does not disclose each and every feature required by claims 1 and 29 and consequently does not anticipate any of claims 1 or 29.

The Howard patent describes preparation of a low tack glue that incorporates polymeric microspheres. (Howard patent: line 67, column 1, through line 3, column 2). The Howard patent emphasizes the polymeric microspheres "are derived from non-ionic monomers." (Howard patent: lines 3-6, column 2). The Howard patent says a suspension stabilizer and a cationic emulsifier are used during the polymerization technique employed to form the polymeric microspheres. (Howard patent: lines 6-8, column 2).

The Howard patent, with reference to U.S. Patent No. 4,166,152 of Baker et. al, states:

The anionic emulsifiers retard particle coalescence [in a composition containing microspheres (i.e. particles)] by surrounding the particle with a negatively charged double layer which provides repulsion of the individual particles.

(Howard patent: lines 24-27, column 1; clarifying comments in brackets added). The Howard patent then mentions preparation of a low tack glue containing microspheres, where the glue is prepared using a cationic emulsifier, as opposed to the anionic emulsifier disclosed in the Baker patent, so a cationic charge is associated with the microspheres in the glue of the Howard patent. (Howard patent: lines 55-58 and 39-42, column 1). Thus, the Howard patent is concerned with forming a glue containing microspheres that are surrounded by a positively charged layer (instead of the negatively charged layer of the Baker patent) by virtue of using a cationic emulsifier in the glue (as opposed to the anionic emulsifier of the Baker patent) to retard particle

(microsphere) coalescence. This last point is confirmed by the following language employed in claim 1 of the Howard patent: “wherein the microspheres are surrounded by a cationic surfactant which prevents particle coalescence.” (Howard patent: lines 15-17, column 4).

Thus, though the Howard patent employs the language “cationic microspheres” (line 46, column 1) and talks about “cationically (positively) charged” microspheres (lines 40-41, column 1), it is clear the Howard patent is not actually producing microspheres that incorporate a structural cationic charge. Instead, the microspheres of the Howard patent, while in the glue composition, are surrounded by the cationic (positively charged) surfactant so that the combination of the microspheres and the surrounding surfactant have a net cationic charge.

Claim 1 of the above-identified application reads as follows:

1. *(Original) An adhesive composition, the adhesive composition comprising:
a plurality of cationic microspheres; and
a fluidizing medium effective for supporting fluid application of the
adhesive composition to a surface.*

Thus, claim 1 defines an adhesive composition that comprises a fluidizing medium and cationic microspheres. Next, claim 29 of the above-identified application reads as follows:

29. *(Original) A method of making an adhesive composition, the method comprising:
causing a plurality of cationic microspheres to exist in a fluidizing
medium to yield the adhesive composition, the fluidizing medium
effective for supporting fluid application of the adhesive
composition to a surface.*

Thus, claim 29 defines a method of making an adhesive composition that concerns a fluidizing medium and cationic microspheres. The above-identified application makes it very clear the cationic microspheres defined in claims 1 and 29 are very different from the microspheres employed in the Howard patent.

The Examiner seeks to turn Applicants’ explanation distinguishing the cationic microspheres of claims 1 and 29 from the combination of microspheres and cationic surfactant disclosed in the Howard patent:

[A]pplicants appear to admit at page 19, lines 6-8 and at page 20, lines 11-13 that the reference discloses microspheres that one of ordinary skill in the art would consider to be ‘cationic.’

into an admission. First complete paragraph of Office Action dated January 30, 2006, referring to Amendment filed December 1, 2006. The Howard microspheres are not the cationic microspheres defined in the present application, such as in claims 1 and 29. The Examiner is directed to the text referenced by the Examiner in Applicants' Amendment, which says there is a cationic charge associated with the Howard microspheres due to the cationic surfactant that surrounds the Howard microspheres. This is far different from the Examiner's contention about the Howard microspheres being cationic. The Howard patent clearly does not disclose anything about the Howard microspheres being cationic, but instead discloses use of a different component, the cationic surfactant, that associates a cationic charge with the microspheres so long as the cationic surfactant is present.

The Examiner seeks to rely solely on the "cationic microsphere" claim language without considering the true meaning of the "cationic microsphere" term, as recited in claims 1 and 29 of the above-identified application:

More particularly, applicants argue (e.g. page 17, 1st complete paragraph) that the 'Howard patent is not actually producing microspheres that incorporate a structural cationic charge'. However, the Examiner submits that the reference either expressly or inherently discloses applicants' straightforwardly claimed (e.g. claim 1) 'cationic microspheres The fact that these cationic microspheres may be formed in a significantly different manner than those of applicants' invention is patentably irrelevant in view of their broad, chosen claim language.

First complete paragraph on page 2 of Office Action dated January 30, 2006.

The Examiner's attempt to disregard the specification when determining the meaning of the "cationic microsphere" term, as recited in claims 1 and 29, is erroneous. During examination, "claims . . . are to be given their broadest reasonable interpretation consistent with the specification, and . . . claim language should be read in light of the specification as it would be interpreted by one of ordinary skill in the art." In re American Academy of Science Tech Center, 70 USPQ2d 1827, 1834 (Fed. Cir. 2004) (emphasis added) (citing In re Bond, 15 USPQ2d 1566, 1567 (Fed. Cir. 1990)). "Although the PTO must give claims their broadest reasonable interpretation, this interpretation must be consistent with the one that those skilled in the art would reach." In re American Academy of Science Tech Center, 70 USPQ2d at 1834 (citing In re Cortright, 49 USPQ2d 1464, 1468 (Fed. Cir. 1999)). "[T]he PTO must apply the

broadest reasonable meaning to the claim language, taking into account any definitions presented in the specification.” In re American Academy of Science Tech Center, 70 USPQ2d at 1834 (Fed. Cir. 2004) (citing In re Bass, 65 USPQ2d 1156 (Fed. Cir. 2002)).

The specification of the above-identified application does explain what is meant by the “cationic microsphere” term that is recited in claims 1 and 29. First, the application states the cationic microspheres are “a reaction product of certain polymerizable substances:”

Cationic polymeric elastomeric microspheres (also referred to herein as “cationic microspheres”) that are produced in accordance with the present invention are a reaction product of certain polymerizable substances.

(Lines 6-8, page 3, of the above-identified application). The cationic charge of the Howard microspheres, on the other hand, results from employing a cationic emulsifier in the Howard glue, such that the microspheres of the Howard glue are surrounded by a positively charged layer, by virtue of incorporating the cationic emulsifier in the glue to retard particle (microsphere) coalescence. (Howard patent: lines 55-58 and 39-42, column 1). Again, this last point is confirmed by the following language employed in claim 1 of the Howard patent: “wherein the microspheres are surrounded by a cationic surfactant which prevents particle coalescence.” (Howard patent: lines 15-17, column 4). Thus, the cationic microspheres of the present invention, as defined in claims 1 and 29, are “a reaction product of certain polymerizable substances,” whereas the microspheres of the Howard patent bear a positive charge because the Howard microspheres are surrounded by a cationic surfactant.

Another distinction is the reaction mixture employed to form adhesive composition that comprises the cationic microspheres of the present application may, but does not necessarily, include a cationic surfactant:

The polymerizable substances are included as part of a reaction mixture in the course of preparing the cationic microspheres. In addition to the polymeric substances, the reaction mixture also includes one or more catalyzation initiators. The reaction mixture will typically also include a fluidizing medium, such as deionized water, and a surfactant, such as a cationic surfactant.

(Lines 13-17, page 3, of the above-identified application). The Howard patent, on the other hand, necessarily employs a cationic surfactant (emulsifier) during preparation of the low tack glue that comprises the Howard microspheres. (Howard patent: lines 6-8, column 2).

Finally, the present application states the cationic microspheres are cationically charged. The following details about this cationic charge of the cationic microspheres are provided in the present application:

. . . . Furthermore, the cationic microspheres included in the repositionable PSA have a cationic charge. . . . It is believed the chemical component of the cationic microspheres that causes the cationic microspheres to carry the cationic charge is a structural portion of the cationic microspheres The cationic microspheres are therefore believed to be unsusceptible to losing the cationic charge.

(Lines 18-26, page 18, of the above-identified application). Thus, the present application states a chemical component of the cationic microspheres that causes the cationic microspheres to carry the cationic charge is believed to be a structural portion of the cationic microspheres and that the cationic microspheres are believed to be unsusceptible of losing this structural cationic charge. The Howard patent, on the other hand, includes no such structural component that contributes a cationic charge to the Howard microspheres. Instead, the Howard microspheres have an associated cationic charge due to the cationic surfactant which surrounds the microspheres and thereby "prevents particle coalescence." (Howard patent: lines 15-17, column 4).

Furthermore, the doctrine of claim differentiation demonstrates the microspheres of the Howard patent are distinct and different from the cationic microspheres of claims 1 and 29. The doctrine of claim differentiation arises from "the common sense notion that different words or phrases used in separate claims are presumed to indicate that the claims have different meanings and scope." Karlin Tech. Inc. v. Surgical Dynamics, Inc., 50 USPQ2d 1465, 1468 (Fed. Cir. 1999). The doctrine of claim differentiation supports a broader construction of an independent claim versus a claim that depends from the independent claim because the doctrine creates a rebuttable presumption that each claim has a different scope. See Karlin Tech. at 1468. Thus, according to the doctrine, the features recited in a dependent claim should ordinarily not be read into an independent claim that is the basis of the dependent claim. Indeed, the presumption of the doctrine is at its strongest "where the limitation sought to be 'read into' an independent

claim already appears in a dependent claim," Liebel-Flarsheim Co. v. Medrad, Inc., 69 USPQ2d 1801, 1807 (Fed. Cir. 2004).

The doctrine of claim differentiation leads to the conclusion the cationic microspheres defined in claims 1 and 29 are not disclosed in the Howard patent. Any conclusion to the contrary would cause claim 7 to be meaningless. The microspheres of the Howard patent have no associated cationic charge absent placement of cationic surfactant about the Howard microspheres. As noted below, claim 7 depends from claim 1 that requires "cationic microspheres." Claim 7, by virtue of depending from claim 1, defines an adhesive composition that requires both "cationic microspheres" and "a cationic surfactant." If the combination of the microspheres of the Howard patent and the cationic surfactant of the Howard patent were considered to equal the cationic microspheres of claims 1 and 29 of the present application, claim 7 would wind up adding nothing to claim 1 of the present application. This is clearly an absurd result, particularly considering the structural charge incorporation in the cationic surfactant taught by the present application. The doctrine of claim differentiation presumes the cationic surfactant defined in claim 7 adds something in addition to the cationic microsphere defined in claims 1 and 29, absent evidence to the contrary. See Karlin Tech. at 1468. The Examiner has not produced any such evidence to the contrary, so the result of application of the doctrine of claim differentiation is believed valid and probative.

The foregoing comments demonstrate that one of ordinary skill in the art would understand the cationic microspheres recited in claims 1 and 29 of the above-identified application are distinct and different from the microspheres employed in the Howard patent. Consequently, it is clear the Howard patent fails to disclose the cationic microspheres defined in claims 1 and 29 of the present application. Therefore, the Howard patent fails to disclose each and every detail of either claim 1 or claim 29, as required by Bond, 15 U.S.P.Q.2d at 1567. Thus, the Howard patent does not anticipate either claim 1 or claim 29.

Claims 1 and 29 are believed allowable, since the Howard patent does not disclose each and every detail required by claim 1 or claim 29. Consequently, the Examiner erred in rejecting claims 1 and 29 under U.S.C. §102(b) based upon the Howard patent.

Reconsideration and reversal of the rejections of claims 1 and 29 accompanied by allowance of claims 1 and 29 are respectfully requested.

B. The Examiner failed to establish that the Howard patent discloses each and every element of claim 7.

The Examiner has also alleged the Howard patent anticipates claim 7 under 35 U.S.C. §102(b). However, despite the Examiner's statements in support thereof, the Howard patent does not in fact anticipate claim 7 under 35 U.S.C. §102(b), since "every element of the claimed invention must be identically shown in a single reference" if a prior art reference is to anticipate in terms of 35 U.S.C. §102. In re Bond, 15 U.S.P.Q.2d at 1567.

As noted above in relation to addressing the Examiner's rejection of claims 1 and 29 based on the Howard patent, the microspheres of the Howard include no such structural component that contributes a cationic charge to the Howard microspheres. Instead, the Howard microspheres have an associated cationic charge due to the cationic surfactant which surrounds the microspheres and thereby "prevents particle coalescence." (Howard patent: lines 15-17, column 4). On the other hand, as explained above, the cationic microspheres of claims 1 and 29 are believed to include a chemical component as a structural portion of the cationic microspheres that causes the cationic microspheres to carry the cationic charge and renders the cationic microspheres unsusceptible of losing this structural cationic charge.

Claim 7 depends from claim 1 that requires "cationic microspheres." Claim 7 reads as follows:

7. *(Original) The adhesive composition of claim 1 wherein the adhesive composition further comprises a cationic surfactant.*

Thus, claim 7 defines an adhesive composition that requires both "cationic microspheres" and "a cationic surfactant."

The Howard patent, as explained above in relation to addressing the Examiner's rejection of claims 1 and 29 based on the Howard patent, does not disclose both cationic microspheres and a cationic surfactant. Instead, the Howard patent discloses a combination of microspheres and a cationic surfactant. The Howard microspheres have an associated cationic

charge due to the cationic surfactant which surrounds the microspheres and thereby “prevents particle coalescence.” (Howard patent: lines 15-17, column 4). However, absent this cationic surfactant, the Howard microspheres would have no associated cationic charge.

Furthermore, as explained above, the doctrine of claim differentiation presumes the cationic surfactant defined in claim 7 adds something in addition to the cationic microsphere defined in claims 1 and 29, absent evidence to the contrary. See Karlin Tech. at 1468. The Examiner has not produced any such evidence to the contrary, so the result of application of the doctrine of claim differentiation is believed valid and probative.

The foregoing comments demonstrate to disclose both the cationic surfactant and the cationic microspheres that claim 7 requires. Therefore, the Howard patent fails to disclose each and every detail of claim 7, as required by Bond, 15 U.S.P.Q.2d at 1567. Thus, the Howard patent does not anticipate claim 7.

Claim 7 is believed allowable, since the Howard patent does not disclose each and every detail required by claim 7. Consequently, the Examiner erred in rejecting claim 7 under U.S.C. §102(b) based upon the Howard patent. Reconsideration and reversal of the rejection of claim 7 accompanied by allowance of claim 7 is respectfully requested.

C. Claims 4, 8-10, and 36-37 each depend from an allowable claim and are each therefore allowable.

The Examiner has alleged the Howard patent anticipates claims 4, 8-10, and 36-37 under 35 U.S.C. §102(b). Claims 4, 8-10 each depend from allowable independent claim 1. Since claims 4, 8-10 each depend from independent claim 1, the Examiner has failed to establish that the inventions of claims 4, 8-10 are anticipated by the Howard patent, for the reasons stated above with respect to claim 1. Consequently, the Examiner erred in rejecting claims 4, 8-10 under U.S.C. §102(b) based upon the Howard patent. Claims 36-37 each depend from allowable independent claim 29. Since claims 36-37 each depend from independent claim 29, the Examiner has failed to establish that the inventions of claims 36-37 are anticipated by the Howard patent, for the reasons stated above with respect to claim 29. Consequently, the

Examiner erred in rejecting claims 4, 8-10, and 36-37 under U.S.C. §102(b) based upon the Howard patent. Reconsideration and reversal of the rejections of claims 4, 8-10, and 36-37 accompanied by allowance of claims 4, 8-10, and 36-37 are respectfully requested.

I. (b). The Howard Patent Does Not Teach, Suggest, Disclose, or Render Obvious The Present Invention, As Defined In Claims 1, 4, 7-10, 29, 36 and 37.

A. The Examiner failed to establish a prima facie case with respect to the alleged obviousness of claims 1, 7, and 29.

Under 35 U.S.C. §103, the Examiner bears the burden of establishing a *prima facie* case of obviousness. In re Rijckaert, 28 U.S.P.Q.2d 1955, 1956 (Fed. Cir. 1993). To establish a *prima facie* case of obviousness, the Examiner must show the teachings from the prior art would appear to have suggested the claimed subject matter to a person of ordinary skill in the art. Rijckaert, 28 U.S.P.Q.2d at 1956. To establish *prima facie* obviousness, the Examiner must show that the prior art **compels** the conclusion the Examiner seeks, and not merely that the prior art would reasonably allow the conclusion the Examiner seeks. In re Spada, 15 U.S.P.Q.2d 1655, 1657, n. 3 (Fed. Cir. 1990). "If the Examiner fails to establish a *prima facie* case, the rejection is improper and will be overturned." Rijckaert, 28 U.S.P.Q.2d at 1956.

The Examiner has never made any allegations about details recited in claims 1, 7, or 29 allegedly being obvious considering the Howard patent, standing alone. Instead, the Examiner has alleged only that the details recited in claims 1 and 29 are disclosed by the Howard patent. Interestingly, the Examiner has never even alleged anything about the details recited in claim 7. The foregoing observations demonstrate the Examiner has not met his burden of establishing a *prima facie* case of obviousness pursuant to Rijckaert. Since the Examiner has failed to establish a *prima facie* case, the rejection of claims 1, 7, and 29 under 35 U.S.C. §103 based on the Howard patent is improper and should be overturned, in accordance with the guidance from Rijckaert. Consequently, Applicants respectfully request that the Examiner's rejection of claims 1, 7, and 29 under 35 U.S.C. §103(a) be reversed and that claims 1, 7, and 29 be allowed.

B. The Howard patent fails to teach, suggest, disclose, or render obvious each and every element of claims 1, 7, and 29.

The Examiner has alleged the Howard patent renders claims 1, 7 and 29 obvious under 35 U.S.C. §103(a). However, despite the Examiner's statements in support thereof, the Howard patent does not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 1, 7 and 29.

Some teaching, suggestion, or motivation must exist to modify a prior art reference in the fashion suggested or detailed by an Examiner. The mere fact that a reference could be modified to produce the claimed invention does not make the claimed invention obvious. Libbey-Owens Ford Co. v. BOC Group Inc., 4 U.S.P.Q.2d 1097, 1103 (D.N.J. 1987). Furthermore, the mere fact that the prior art discloses the components of a claimed invention does not make the invention obvious. Northern Telecom, Inc. v. Datapoint Corp., 15 U.S.P.Q.2d 1321, 1323 (Fed. Cir. 1990). Instead, "[s]omething in the prior art as a whole must suggest the desirability, and thus the obviousness, of making the combination." Uniroyal, Inc. v. Rudkin-Wiley Corp., 5 U.S.P.Q.2d 1434, 1438 (Fed. Cir. 1988).

As noted above, the Examiner has never made any allegations about details recited in claims 1, 7, or 29 allegedly being obvious considering the Howard patent, standing alone. Instead, the Examiner has alleged only that the details recited in claims 1 and 29 are disclosed by the Howard patent, but has not ever alleged anything about the details recited in claim 7.

The Howard patent describes preparation of a low tack glue that incorporates polymeric microspheres. (Howard patent: line 67, column 1, through line 3, column 2). The Howard patent emphasizes the polymeric microspheres "are derived from non-ionic monomers." (Howard patent: lines 3-6, column 2). The Howard patent says a suspension stabilizer and a cationic emulsifier are used during the polymerization technique employed to form the polymeric microspheres. (Howard patent: lines 6-8, column 2). As noted above in relation to the Examiner's anticipation rejections based on the Howard patent, the Howard patent concerns formation of a glue containing microspheres that are surrounded by a positively charged layer

(instead of the negatively charged layer of the Baker patent) by virtue of using a cationic emulsifier in the glue (as opposed to the anionic emulsifier of the Baker patent) to retard particle (microsphere) coalescence. This last point is confirmed by the following language employed in claim 1 of the Howard patent: "wherein the microspheres are surrounded by a cationic surfactant which prevents particle coalescence." (Howard patent: lines 15-17, column 4).

As noted previously, claim 1 defines an adhesive composition that comprises a fluidizing medium and cationic microspheres and claim 29 defines a method of making an adhesive composition that concerns a fluidizing medium and cationic microspheres. Claim 7 depends from claim 1 and further requires a cationic surfactant.

As established above in relation to the Examiner's anticipation rejection of claims 1 and 29, claims 1 and 29 require cationic microspheres produced as a result of polymerization of polymerizable substances, where the structure of the cationic microspheres carries the cationic charge. The cationic microspheres of claims 1 and 29 are not susceptible to losing the cationic charge. On the other hand, the Howard microspheres are based on non-ionic monomers, so the cationic charge associated with the Howard microspheres is associated with the microspheres, but is not a part of the structure of the Howard microspheres.

In essence, the microspheres produced as a result of the polymerization of polymerizable substances in accordance with Howard do not result in any cationic charge association with the Howard microspheres. Rather, the cationic charge associated with the Howard microspheres comes about due to mixture of the Howard microspheres with the cationic surfactant. Thus, as explained previously, the Howard patent does not disclose the cationic microspheres defined in claims 1 and 29, but instead discloses a combination of non-ionic microspheres and cationic surfactant. The cationic surfactant shares cationic charge with the non-ionic microspheres, but there is no indication, suggestion, or disclosure that combination of the cationic surfactant and Howard microspheres are the same as the cationic microspheres defined in claims 1 and 29. Furthermore, as demonstrated with reference to the doctrine of claim differentiation, inclusion of the cationic surfactant detail in claim 7 further highlights the

distinction between the microsphere/cationic surfactant combination of the Howard patent and the cationic microspheres defined in claims 1 and 29.

Thus, it is clear the Howard patent does not actually disclose or produce microspheres that incorporate a structural cationic charge. Instead, the microspheres of the Howard patent, while in the glue composition, are surrounded by the cationic (positively charged) surfactant so the combination of the microspheres and the surrounding surfactant have a net cationic charge. On the other hand, the cationic microspheres of claims 1 and 29 bear a structural charge, and may be produced and utilized in the absence of any cationic surfactant. Simply said, the microspheres of the Howard patent are different from, and do not disclose, the cationic microspheres of claims 1 and 29. Furthermore, the microsphere/cationic surfactant combination of the Howard patent is different from, and does not disclose, the cationic microspheres of claims 1 and 29. Finally, the combination of the cationic microspheres and cationic surfactant required by claim 7 is different from both the microspheres of the Howard patent and is different from the microsphere/cationic surfactant combination of the Howard patent.

Furthermore, there is no teaching, suggestion or motivation evident from the Howard patent to somehow modify the microspheres of the Howard patent or the microsphere/cationic surfactant combination of the Howard patent to equal the cationic microspheres defined in claims 1 and 29. Likewise, there is no teaching, suggestion or motivation evident from the Howard patent to somehow modify the microspheres of the Howard patent so the Howard microspheres would, like the cationic microspheres of the present application, be cationically charged so the Howard cationic surfactant could optionally be eliminated. Thus, the Howard patent lacks any teaching or suggestion leading to the combination of cationic surfactant and cationic microspheres required by claim 7. Indeed, the Examiner does not mention anything about any such modifications. That is likely due to the fact that there is no teaching or suggestion in the Howard patent to modify the microspheres of the Howard patent so the cationic surfactant can be removed from the Howard recipe.

Lacking any such teaching or suggestion, it is clear, pursuant to Libbey-Owens Ford, the Howard patent does not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 1, 7, and 29. Consequently, Applicants believe the Examiner erred in rejecting claims 1, 7, and 29 under U.S.C. §103(a) based upon the Howard patent and that claims 1, 7, and 29 are allowable. Reconsideration and reversal of the rejections of claims 1, 7, and 29 accompanied by allowance of claims 1, 7, and 29 are respectfully requested.

C. Claims 4, 8-10, and 36-37 each depend from an allowable claim and are each therefore allowable.

The Examiner has alleged the Howard patent renders claims 4, 8-10, 36, and 37 obvious under 35 U.S.C. §103(a). Claims 4, 8-10 each depend from allowable independent claim 1. Since claims 4, 8-10 each depend from independent claim 1, the Examiner has failed to establish that the inventions of claims 4, 8-10 are rendered obvious by the Howard patent, for the reasons stated above with respect to claim 1. Consequently, the Examiner erred in rejecting claims 4, 8-10 under U.S.C. §103(a) based upon the Howard patent. Claims 36-37 each depend from allowable independent claim 29. Since claims 36-37 each depend from independent claim 29, the Examiner has failed to establish that the inventions of claims 36-37 are rendered obvious by the Howard patent, for the reasons stated above with respect to claim 29. Consequently, the Examiner erred in rejecting claims 4, 8-10, and 36-37 under U.S.C. §103(a) based upon the Howard patent. Reconsideration and reversal of the rejections of claims 4, 8-10, and 36-37 accompanied by allowance of claims 4, 8-10, and 36-37 are respectfully requested.

II. The Howard and Crandall Patents Do Not Teach, Suggest, or Make Obvious the Invention of the Present Application as Defined in Claims 1-51.

The Examiner has alleged the Howard and Crandall patents render claims 1-51 obvious under 35 U.S.C. §103(a). In support of this rejection, the Examiner initially provided the following comments:

Claims 1-51 are rejected under 35 U.S.C. 103(a) as being unpatentable over Howard combined with Crandall et al. Howard is again relied upon substantially as set forth, above, while Crandall et al discloses (note particularly the Abstract, Col 1, lines 10-19, Col 3, line 43 – Col 4, line 37, Col 6, line 59 – Col 9, line 54, Col 10, lines 26 - 36, Examples) what essentially appears to comprise applicants' broad genus of cationic adhesive microspheres that are suitable for forming the claimed genus of repositionable adhesive compositions, together with the accompanying methods of making and using, various adhesive compositions, coated articles and the like. The references are clearly combinable, each featuring repositionable adhesive compositions which feature cationic microspheres, with the microspheres of Crandall et al improving the adhesive compositions of Howard by incorporating their microspheres in place of the Howard microspheres, motivated by (Col 3, lines 44 - 47) an expectation of enhanced stability and performance properties, while maintaining inherent tackiness, elastomeric properties and solvent or water dispersibility. Other parameters that are not either expressly or inherently disclosed are each believed to be routine optimizations to one of ordinary skill, in the absence of unexpected results.

Office Action dated September 1, 2005. Despite these comments, the Howard patent and the Crandall patent, either separately or in combination, fail to teach, suggest, or make obvious the invention of the above-identified application, as defined in claims 1-51.

The Crandall patent discloses microspheres based optionally in part on a comonomer, where the optional comonomer may be nonpolar, ionic, polar, or mixtures thereof. (Crandall patent: lines 59-63, column 6, and lines 1-2, column 7). Examples of suitable comonomers are provided. (Crandall patent: line 13, column 8, through line 50, column 9). However, no teaching or example of a cationic unsaturated vinyl comonomer is provided anywhere within the text, examples, or claims of the Crandall. Indeed, the Examiner did not even allege the Crandall patent teaches, suggests, or discloses the cationic unsaturated vinyl comonomer(s) disclosed and claimed in the above-identified application. Furthermore, the

Examiner did not identify any particular optional comonomer disclosed in the Crandall patent as an example of the cationic unsaturated vinyl comonomer(s) disclosed and claimed in the above-identified application. Instead, the Examiner chose to rely on the broad, conclusory contention about the Crandall patent allegedly disclosing "what essentially appears to comprise applicants' broad genus of cationic adhesive microspheres." Despite this allegation, Applicants assert the Crandall patent does not teach, suggest, disclose, or make obvious the cationic unsaturated vinyl comonomer(s) disclosed and claimed in the above-identified application.

In response to Applicant's argument included in the Amendment filed December 1, 2006, the Examiner responded as follows regarding the cationic unsaturated vinyl monomer missing from the Crandall patent:

The Examiner disagrees with applicants' contention (Response, page 22, 1st complete paragraph) that Crandall et al fails to teach, suggest or disclose a 'cationic unsaturated vinyl monomer.' Note, e.g. Col 3, lines 43-47 which clearly teaches such a genus of embodiments, which are also believed to be capable of being polymerized and thus become part of the microspheres as clearly implied by the aforementioned citation, applicants' comments (e.g. Response, bridging paragraph, pages 22-23) to the contrary notwithstanding.

Office Action dated January 30, 2006; paragraph extending from the bottom of page 2 to the top of page 3. The passage relied upon by the Examiner from col. 3, lines 43-47, of the Crandall patent read as follows:

It has now been discovered that microspheres that contain vinyl unsaturated additives with both an ionic and hydrophobic moiety to enhance stability and performance properties, while maintaining inherent tackiness, elastomeric properties and solvent or water dispersibility.

While this passage does appear to pertain to ionic vinyl unsaturated additives, the ionic genus does not necessarily teach cationic vinyl unsaturated additives. More of a teaching is needed, but the Examiner has not pointed to where such additional teaching may be found in the Crandall patent. Thus, the Examiner continues to rely on the broad, conclusory contention about the Crandall patent allegedly disclosing "what essentially appears to comprise applicants' broad genus of cationic adhesive microspheres," even though it actually discloses an ionic genus, rather than a cationic species or genus. Therefore, despite this further allegation, Applicants assert the

Crandall patent does not teach, suggest, disclose, or make obvious the cationic unsaturated vinyl comonomer(s) disclosed and claimed in the above-identified application.

The Crandall patent also discloses incorporation of a vinyl unsaturated additive along with the polymerizable components that are polymerized to form the microspheres. (Crandall patent: lines 59-66, column 6). Examples of suitable vinyl unsaturated additives are provided. (Crandall patent: line 43, column 7, through line 12, column 8). There is no disclosure in the Crandall patent about the vinyl unsaturated additive being polymerized to form part of the microsphere. Furthermore, the Examiner has not alleged the vinyl unsaturated additive is polymerized to form part of the microsphere. Instead, the Crandall patent teaches the vinyl unsaturated additive helps prevent the microspheres that are formed from coagulating during synthesis. (Crandall patent: lines 16-27, column 16). Further evidence that the vinyl unsaturated additive is not polymerized to form part of the microsphere is found in claim 1 of the Crandall patent. Claim defines a “stabilized microsphere adhesive composition” that comprises, among other elements, “a plurality of polymeric, elastomeric microspheres . . .” and the vinyl unsaturated additive. (Crandall patent: lines 39-51, column 18). Thus, the Crandall patent does not teach, suggest, or disclose anything about the vinyl unsaturated additive being polymerized to form part of the microsphere. Instead, the Crandall patent teaches, suggests, and discloses the vinyl unsaturated additive is not polymerized and does not form part of the microsphere.

A. The Howard and Crandall patents fail to teach, suggest, disclose, or render obvious each and every element of claims 1, 18, and 29.

The Examiner has alleged the Howard and Crandall patents render claims 1, 18 and 29 obvious under 35 U.S.C. §103(a). Claims 1, 18, and 29 each recite cationic microspheres. Despite the Examiner’s statements in support thereof, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 1, 18 and 29.

Some teaching, suggestion, or motivation must exist to modify a prior art reference in the fashion suggested or detailed by an Examiner. The mere fact that a reference

could be modified to produce the claimed invention does not make the claimed invention obvious. Libbey-Owens Ford Co. v. BOC Group Inc., 4 U.S.P.Q.2d 1097, 1103 (D.N.J. 1987). Furthermore, the mere fact that the prior art discloses the components of a claimed invention does not make the invention obvious. Northern Telecom, Inc. v. Datapoint Corp., 15 U.S.P.Q.2d 1321, 1323 (Fed. Cir. 1990). Instead, "[s]omething in the prior art as a whole must suggest the desirability, and thus the obviousness, of making the combination." Uniroyal, Inc. v. Rudkin-Wiley Corp., 5 U.S.P.Q.2d 1434, 1438 (Fed. Cir. 1988).

As noted above, claims 1, 18, and 29 each recite cationic microspheres. The Examiner suggests that cationic microspheres are disclosed in the Crandall patent and could be substituted in place of the non-ionic microspheres disclosed in the Howard patent. This allegation is troubling for at least one major reason. The present application defines the cationic microspheres of the present invention as being based on a cationic unsaturated vinyl monomer; such a cationic unsaturated vinyl monomer is neither taught nor disclosed in the Crandall patent. Furthermore, Applicants find no suggestion in either the Crandall patent nor the Howard patent to substitute the alleged cationic microspheres of the Crandall patent in place of the non-ionic microspheres of the Howard patent, even if Crandall did (it does not) teach preparation of cationic microspheres. The Examiner points to no particular language in either the Howard or Crandall patent, but instead relies on the misplaced theory that Howard and Crandall both teach cationic microspheres so that provides motivation to swap microspheres. This theory is problematic at a basic level, since neither the Howard patent (see prior discussions of anticipation and obviousness rejections based on Howard alone) nor the Crandall patent (see above) teach preparation of cationic microspheres.

The specification of the above-identified application explains what is meant by the "cationic microsphere" term that is recited in claims 1, 18, and 29. First, the application states the cationic microspheres are derived, in part, from one or more cationic unsaturated vinyl comonomers:

Cationic polymeric elastomeric microspheres (also referred to herein as "cationic microspheres") that are produced in accordance with the present invention are a reaction product of certain polymerizable substances. The polymerizable

substances include at least one C₄-C₁₄ alkyl (meth) acrylate monomer and one or more cationic unsaturated vinyl comonomers.

(Lines 6-10, page 3, of the above-identified application; emphasis added). From the discussion above, it has been established that the “cationic microspheres” term of claims 1, 18, and 29 means microspheres that include a cationically-charged chemical component as a structural portion of the microspheres.

As pointed out previously in regard to the Examiner’s rejection based solely on the Howard patent, there is no teaching, suggestion, or motivation based on the disclosure of the Howard patent to include a cationically-charged chemical component as a structural portion of the Howard microspheres, as required by the cationic microspheres recited in claims 1, 18, and 29. The Crandall patent, as noted above, does not teach, suggest, or disclose a polymerizable cationic unsaturated vinyl comonomer. Instead, the Crandall patent merely teaches polymerizable ionic, but not cationic, comonomers. (Crandall patent: lines 59-63, column 6, and lines 1-2, column 7). Consequently, there is no teaching, suggestion, or motivation based on the disclosure of the Crandall patent to select and include a cationically-charged chemical component as a structural portion of the Crandall microspheres, as required by the cationic microspheres recited in claims 1, 18, and 29. No teaching, suggestion, or motivation, as required by Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103, exists to modify the Crandall patent in the fashion suggested or detailed by an Examiner. The mere fact that a reference could be modified to produce the claimed invention does not make the claimed invention obvious. Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103.

The only conceivable basis for the Examiner’s apparent suggestion of deriving the Crandall microsphere from a cationically-charged chemical component that is included as a structural portion of the Crandall microspheres that contributes the cationic charge in the cationic microspheres and is permanently and chemically bound within the various Crandall microspheres, per claims 1, 18, and 29, would be to rely on the teachings of the specification of the present application as a road map via hindsight reconstruction. However, it is well known that such hindsight reconstruction is an improper basis for alleging obviousness. Hindsight reconstruction of a claimed invention using the claimed invention as a model for picking and choosing details to use in modifying a prior art

disclosure is improper. See In re Fine, 5 U.S.P.Q.2d 1596, 1600 (Fed. Cir. 1988). An Examiner should forget the invention at issue when considering whether it would be obvious to modify a prior art disclosure. See W.L. Gore & Assoc., Inc. v. Garlock, Inc., 220 U.S.P.Q. 303, 312-13 (Fed. Cir. 1983).

Thus, the Examiner's rejection based on the Howard patent and the Crandall patent fails. Even if the microspheres of the Crandall patent were substituted in place of the Howard patent, per the Examiner's suggestion, the resulting combination would still fail to include a cationically-charged chemical component as a structural portion of the microspheres, as required by the cationic microspheres recited in claims 1, 18 and 29.

Lacking the required teaching and suggestion to make the modifications suggested by the Examiner, it is clear, pursuant to Libbey-Owens Ford, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 1, 18, and 29. Consequently, Applicants believe the Examiner erred in rejecting claims 1, 18, and 29 under U.S.C. §103(a) based upon the Howard and Crandall patents and that claims 1, 18, and 29 are allowable. Reconsideration and reversal of the rejections of claims 1, 18, and 29 accompanied by allowance of claims 1, 18, and 29 are respectfully requested.

B. The Howard and Crandall patents fail to teach, suggest, disclose, or render obvious each and every element of claims 3, 19, and 38.

The Examiner has alleged the Howard and Crandall patents render claims 3, 11, 19, and 38 obvious under 35 U.S.C. §103(a). Claims 3, 11, 19, and 38 each recite a cationic unsaturated vinyl monomer. Despite the Examiner's statements in support thereof, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 3, 11, 19 and 38.

Each of claims 3, 11, 19, and 38 define a cationic unsaturated vinyl comonomer that is described as being incorporated in cationic microspheres (claim 3), described as a polymerizable substance (claims 11 and 19), described as a component of a polymerized product

(claim 19), or described as part of a polymerizable mixture (claim 38). As explained above, the Crandall patent does not teach, suggest, or disclose a cationic unsaturated vinyl comonomer that is described as a polymerizable substance. Instead, the Crandall patent merely teaches polymerizable ionic, but not cationic, comonomers. (Crandall patent: lines 59-63, column 6, and lines 1-2, column 7).

The Crandall patent thus fails to teach suggest, disclose or make obvious the invention of the above-identified application, as defined in claims 3, 11, 19, and 38. The Examiner's suggestion of substituting the Crandall microspheres in place of the Howard microspheres adds nothing in regard to claims 3, 11, 19, and 38 since neither the Crandall patent nor the Howard patent teaches, suggests, or discloses a cationic unsaturated vinyl comonomer that is described as a polymerizable substance.

As pointed out previously in regard to the Examiner's rejection based solely on the Howard patent, there is no teaching, suggestion, or motivation based on the disclosure of the Howard patent to include a cationically-charged chemical component as a structural portion of the Howard microspheres. The Crandall patent, as noted above, does not teach, suggest, or disclose a polymerizable cationic unsaturated vinyl comonomer. Instead, the Crandall patent merely teaches polymerizable ionic, but not cationic, comonomers. (Crandall patent: lines 59-63, column 6, and lines 1-2, column 7). Consequently, there is no teaching, suggestion, or motivation based on the disclosure of the Crandall patent to select and include a cationically-charged chemical component (i.e. cationic unsaturated vinyl comonomer) as a structural portion of the Crandall microspheres, as recited in claims 3, 11, 19, and 38. No teaching, suggestion, or motivation, as required by Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103, exists to modify the Crandall patent in the fashion suggested or detailed by an Examiner. The mere fact that a reference could be modified to produce the claimed invention does not make the claimed invention obvious. Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103.

The only conceivable basis for the Examiner's apparent suggestion of deriving the Crandall microsphere from a cationically-charged chemical component that is included as a structural portion of the Crandall microspheres, per claims 3, 11, 19, and 38, would be to rely on the teachings

of the specification of the present application as a road map via hindsight reconstruction. However, it is well known that such hindsight reconstruction is an improper basis for alleging obviousness. See In re Fine, 5 U.S.P.Q.2d at 1600.

Thus, the Examiner's rejection based on the Howard patent and the Crandall patent fails. Even if the microspheres of the Crandall patent were substituted in place of the Howard patent, per the Examiner's suggestion, the resulting combination would still fail to include a cationically-charged chemical component as a structural portion of the microspheres, as required by the recitations of claims 3, 11, 19, and 38.

Lacking the required teaching and suggestion to make the modifications suggested by the Examiner, it is clear, pursuant to Libbey-Owens Ford, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 3, 11, 19, and 38. Consequently, Applicants believe the Examiner erred in rejecting claims 3, 11, 19, and 38 under U.S.C. §103(a) based upon the Howard and Crandall patents and that claims 3, 11, 19, and 38 are allowable. Reconsideration and reversal of the rejections of claims 3, 11, 19, and 38 accompanied by allowance of claims 3, 11, 19, and 38 are respectfully requested.

C. The Howard and Crandall patents fail to teach, suggest, disclose, or render obvious each and every element of claims 2 and 30.

The Examiner has alleged the Howard and Crandall patents render claims 2 and 30 obvious under 35 U.S.C. §103(a). Claims 2 and 30 each specify that cationic microspheres have a cationic charge that is permanently and individually bound to each cationic microsphere. Despite the Examiner's statements in support thereof, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 2 and 30.

As explained above, the Crandall patent does not teach, suggest, or disclose a cationic unsaturated vinyl comonomer that is described as a polymerizable substance. Instead,

the Crandall patent merely teaches polymerizable ionic, but not cationic, comonomers. (Crandall patent: lines 59-63 , column 6, and lines 1-2, column 7).

The Crandall patent thus fails to teach suggest, disclose or make obvious the invention of the above-identified application, as defined in claims 2 and 30, since the above-identified application specifies that polymerizable cationic unsaturated vinyl monomers are needed to produce cationic microspheres having a cationic charge, as required by claims 2 and 30, that is permanently and individually bound to each cationic microsphere. The Examiner's suggestion of substituting the Crandall microspheres in place of the Howard microspheres adds nothing in regard to claims 2 and 30 since neither the Crandall patent nor the Howard patent teaches, suggests, or discloses a cationic unsaturated vinyl comonomer that is described as a polymerizable substance.

As pointed out previously in regard to the Examiner's rejection based solely on the Howard patent, there is no teaching, suggestion, or motivation based on the disclosure of the Howard patent to include a cationically-charged chemical component as a structural portion of the Howard microspheres. The Crandall patent, as noted above, does not teach, suggest, or disclose a polymerizable cationic unsaturated vinyl comonomer. Instead, the Crandall patent merely teaches polymerizable ionic, but not cationic, comonomers. (Crandall patent: lines 59-63, column 6, and lines 1-2, column 7). Consequently, there is no teaching, suggestion, or motivation based on the disclosure of the Crandall patent to select and include a cationically-charged chemical component (i.e. cationic unsaturated vinyl comonomer) to yield a cationic charge that is permanently and individually bound to each cationic microsphere, as required by claims 2 and 30. No teaching, suggestion, or motivation, as required by Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103, exists to modify the Crandall patent in the fashion suggested or detailed by an Examiner. The mere fact that a reference could be modified to produce the claimed invention does not make the claimed invention obvious. Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103.

Thus, the Examiner's rejection based on the Howard patent and the Crandall patent fails. Even if the microspheres of the Crandall patent were substituted in place of the Howard patent, per the Examiner's suggestion, the resulting combination would still fail to

include microspheres with cationic charge that is permanently and individually bound to each cationic microsphere, as required by claims 2 and 30.

Lacking the required teaching and suggestion to make the modifications suggested by the Examiner, it is clear, pursuant to Libbey-Owens Ford, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 2 and 30. Consequently, Applicants believe the Examiner erred in rejecting claims 2 and 30 under U.S.C. §103(a) based upon the Howard and Crandall patents and that claims 2 and 30 are allowable. Reconsideration and reversal of the rejections of claims 2 and 30 accompanied by allowance of claims 2 and 30 are respectfully requested.

D. The Howard and Crandall patents fail to teach, suggest, disclose, or render obvious each and every element of claims 5, 13, 22, and 32.

The Examiner has alleged the Howard and Crandall patents render claims 5, 13, 22, and 32 obvious under 35 U.S.C. §103(a). Claims 5, 13, 22, and 32 each define an unsaturated vinyl comonomer. Despite the Examiner's statements in support thereof, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 5, 13, 22, and 32.

Each of claims 5, 13, 22, and 32 define an unsaturated vinyl comonomer that is described as being incorporated in cationic microspheres (claims 5 and 32), described as a polymerizable substance (claims 13 and 22), described as a component of a polymerized product (claim 22), or described as part of a polymerizable mixture (claim 38). As explained above, the Crandall patent does not teach, suggest, or disclose an unsaturated vinyl comonomer that is described as a polymerizable substance.

Specifically, the Crandall patent discloses incorporation of a vinyl unsaturated additive along with the polymerizable components that are polymerized to form the microspheres. (Crandall patent: lines 59-66, column 6). Examples of suitable vinyl unsaturated additives are provided. (Crandall patent: line 43, column 7, through line 12, column 8). There is no disclosure in the Crandall patent about the vinyl unsaturated additive being polymerized to form part of the

microsphere. Furthermore, the Examiner has not alleged the vinyl unsaturated additive is polymerized to form part of the microsphere. Instead, the Crandall patent teaches the vinyl unsaturated additive helps prevent the microspheres that are formed from coagulating during synthesis. (Crandall patent: lines 16-27, column 16). Further evidence that the vinyl unsaturated additive is not polymerized to form part of the microsphere is found in claim 1 of the Crandall patent. Claim defines a “stabilized microsphere adhesive composition” that comprises, among other elements, “a plurality of polymeric, elastomeric microspheres . . .” and the vinyl unsaturated additive. (Crandall patent: lines 39-51, column 18). Thus, the Crandall patent does not teach, suggest, or disclose anything about the vinyl unsaturated additive being polymerized to form part of the microsphere. Instead, the Crandall patent teaches, suggests, and discloses the vinyl unsaturated additive is not polymerized and does not form part of the microsphere.

Thus, there is no teaching, suggestion, or motivation based on the disclosure of the Crandall patent to select and include a polymerizable unsaturated vinyl comonomer as required by claims 5, 13, 22, and 32, to yield a cationic microsphere,. No teaching, suggestion, or motivation, as required by Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103, exists to modify the Crandall patent in the fashion suggested or detailed by an Examiner. The mere fact that a reference could be modified to produce the claimed invention does not make the claimed invention obvious. Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103.

Thus, the Examiner’s rejection based on the Howard patent and the Crandall patent fails. Lacking the required teaching and suggestion to make the modifications suggested by the Examiner, it is clear, pursuant to Libbey-Owens Ford, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 5, 13, 22, and 32. Consequently, Applicants believe the Examiner erred in rejecting claims 5, 13, 22, and 32 under U.S.C. §103(a) based upon the Howard and Crandall patents and that claims 5, 13, 22, and 32 are allowable. Reconsideration and reversal of the rejections of claims 5, 13, 22, and 32 accompanied by allowance of claims 5, 13, 22, and 32 are respectfully requested.

E. The Howard and Crandall patents fail to teach, suggest, disclose, or render obvious each and every element of claims 6, 14, 23, 33, 44, and 47.

The Examiner has alleged the Howard and Crandall patents render claims 6, 14, 23, 33, 44, and 47 obvious under 35 U.S.C. §103(a). Claims 6, 14, 23, 33, 44, and 47 each define a cationic latex adhesive binder. Despite the Examiner's statements in support thereof, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 6, 14, 23, 33, 44, and 47.

The Crandall patent and the Howard patent do not teach, suggest, or disclose a cationic latex adhesive binder. In fact, the Examiner never even mentioned anything about the Crandall patent or the Howard patent and the required cationic latex adhesive binder of claims 6, 14, 23, 33, 44, and 47. Thus, there is no teaching, suggestion, or motivation based on the disclosure of the Crandall patent or the Howard patent to include a cationic latex adhesive binder as required by claims 6, 14, 23, 33, 44, and 47. No teaching, suggestion, or motivation, as required by Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103, exists to modify the Crandall patent in the fashion suggested or detailed by an Examiner.

Thus, the Examiner's rejection based on the Howard patent and the Crandall patent fails. Lacking the required teaching and suggestion to make the modifications suggested by the Examiner, it is clear, pursuant to Libbey-Owens Ford, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 6, 14, 23, 33, 44, and 47. Consequently, Applicants believe the Examiner erred in rejecting claims 6, 14, 23, 33, 44, and 47 under U.S.C. §103(a) based upon the Howard and Crandall patents and that claims 6, 14, 23, 33, 44, and 47 are allowable. Reconsideration and reversal of the rejections of claims 6, 14, 23, 33, 44, and 47 accompanied by allowance of claims 6, 14, 23, 33, 44, and 47 are respectfully requested.

F. The Howard and Crandall patents fail to teach, suggest, disclose, or render obvious each and every element of claims 7, 15, 16, 24, 34, 35, 43, and 45.

The Examiner has alleged the Howard and Crandall patents render claims 7, 15, 16, 24, 34, 35, 43, and 45 obvious under 35 U.S.C. §103(a). Claims 7, 15, 16, 24, 34, 35, 43, and 45 each define a cationic surfactant. As noted previously in relation to claim 7, the Howard patent, standing alone, fails to teach, suggest, disclose, or make obvious use of a cationic surfactant in combination with cationic microspheres of the present invention. Furthermore, despite the Examiner's statements in support thereof, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 7, 15, 16, 24, 34, 35, 43, and 45.

The Examiner relies on the Crandall patent for allegedly teaching production of cationic microspheres and apparently relies on the Howard patent for cationic surfactant that would allegedly remain if Crandall did in fact teach (it does not) preparation of cationic microspheres and substitution of the Crandall microspheres in place of the Howard non-ionic microspheres. However, as explained previously herein at length, the Crandall patent does not teach preparation of cationic microspheres.

Thus, there is no teaching, suggestion, or motivation based on the disclosure of the Crandall patent and the Howard patent to supply cationic surfactant, per claims 7, 15, 16, 24, 34, 35, 43, and 45 along with cationic microspheres, per the Examiner. No teaching, suggestion, or motivation, as required by Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103, exists to modify the Crandall patent in the fashion suggested or detailed by an Examiner and to thereafter substitute the alleged Crandall cationic microspheres in place of the Howard microspheres while retaining the Howard cationic surfactant. The mere fact that a reference could be modified to produce the claimed invention does not make the claimed invention obvious. Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103.

Thus, the Examiner's rejection based on the Howard patent and the Crandall patent fails. Lacking the required teaching and suggestion to make the modifications suggested by the Examiner, it is clear, pursuant to Libbey-Owens Ford, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application,

as defined in claims 7, 15, 16, 24, 34, 35, 43, and 45: Consequently, Applicants believe the Examiner erred in rejecting claims 7, 15, 16, 24, 34, 35, 43, and 45 under U.S.C. §103(a) based upon the Howard and Crandall patents and that claims 7, 15, 16, 24, 34, 35, 43, and 45 are allowable. Reconsideration and reversal of the rejections of claims 7, 15, 16, 24, 34, 35, 43, and 45 accompanied by allowance of claims 7, 15, 16, 24, 34, 35, 43, and 45 are respectfully requested.

G. The Howard and Crandall patents fail to teach, suggest, disclose, or render obvious each and every element of claims 25-26, 46, and 49.

The Examiner has alleged the Howard and Crandall patents render claims 25-26, 46, and 49 obvious under 35 U.S.C. §103(a). Claims 25-26, 46, and 49 each define test results for adhesive produced in accordance with the present invention and subjected to static angle testing, as described in the present application. Despite the Examiner's statements in support thereof, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 25-26, 46, and 49.

Neither the Howard patent nor the Crandall patent teaches, suggests, or disclose use of the test procedures defined in claims 25-26, 46, and 49. To solve this shortcoming, the Examiner uses a broad brushed approach to allege it would be obvious to optimize the alleged combination of the alleged Crandall cationic microspheres in place of the Howard microspheres to obtain the results defined in claims 25-26, 46, and 49:

Other parameters that are not either expressly or inherently disclosed are each believed to be routine optimizations to one of ordinary skill, in the absence of unexpected results.

Office Action dated September 1, 2005. However, the mere fact that a reference could be modified to produce the claimed invention does not make the claimed invention obvious. Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103. Here, the Examiner's allegation is speculative, at best, and there is simply no basis for even believing the Examiner's alleged Crandall cationic microspheres, when substituted in place of the Howard microspheres while retaining the Howard cationic surfactant, per the Examiner, would achieve the claimed results or even possess the claimed cationic microspheres.

Thus, there is no teaching, suggestion, or motivation based on the disclosure of the Crandall patent and the Howard patent to achieve the results defined in claims 25-26, 46, and 49, per the Examiner's suggestion. No teaching, suggestion, or motivation, as required by Libbey-Owens Ford, 4 U.S.P.Q.2d at 1103, exists to modify the Crandall patent in the fashion suggested or detailed by an Examiner and to thereafter substitute the alleged Crandall cationic microspheres in place of the Howard microspheres while retaining the Howard cationic surfactant and then attempting to optimize the combination to achieve the results defined in claims 25-26, 46, and 49.

Thus, the Examiner's rejection based on the Howard patent and the Crandall patent fails. Lacking the required teaching and suggestion to make the modifications suggested by the Examiner, it is clear, pursuant to Libbey-Owens Ford, the Howard and Crandall patents do not teach, suggest, disclose, or render obvious the invention of the above-identified application, as defined in claims 25-26, 46, and 49. Consequently, Applicants believe the Examiner erred in rejecting claims 25-26, 46, and 49 under U.S.C. §103(a) based upon the Howard and Crandall patents and that claims 25-26, 46, and 49 are allowable. Reconsideration and reversal of the rejections of claims 25-26, 46, and 49 accompanied by allowance of claims 25-26, 46, and 49 are respectfully requested.

H. Claims 4, 8-10, 12, 17, 20-21, 27-28, 31, 36-37, 39, 41-42, and 50-51 each depend from an allowable claim and are each therefore allowable.

The Examiner has alleged the Howard and Crandall patents render claims 4, 8-10, 12, 17, 20-21, 27-28, 31, 36-37, 39, 41-42, and 50-51 obvious under 35 U.S.C. §103(a). Claims 4, 8-10 each depend from allowable independent claim 1. Since claims 4, 8-10 each depend from independent claim 1, the Examiner has failed to establish that the inventions of claims 4, 8-10 are rendered obvious by the Howard and Crandall patents, for the reasons stated above with respect to claim 1. Consequently, the Examiner erred in rejecting claims 4, 8-10 under U.S.C. §103(a) based upon the Howard and Crandall patents. Claims 12 and 17 each depend from allowable independent claim 11. Since claims 12 and 17 each depend from independent claim 11, the Examiner has failed to establish that the inventions of claims 12 and 17 are rendered

obvious by the Howard and Crandall patents, for the reasons stated above with respect to claim 11. Claims 20-21 and 27-28 each depend from allowable independent claim 19. Since claims 20-21 and 27-28 each depend from independent claim 19, the Examiner has failed to establish that the inventions of claims 20-21 and 27-28 are rendered obvious by the Howard and Crandall patents, for the reasons stated above with respect to claim 19. Claims 31 and 36-37 each depend from allowable independent claim 29. Since claims 31 and 36-37 each depend from independent claim 29, the Examiner has failed to establish that the inventions of claims 31 and 36-37 are rendered obvious by the Howard and Crandall patents, for the reasons stated above with respect to claim 29. Claims 39, 41-42, and 50-51 each depend from allowable independent claim 38. Since claims 31 and 36-37 each depend from independent claim 38, the Examiner has failed to establish that the inventions of claims 31 and 36-37 are rendered obvious by the Howard and Crandall patents, for the reasons stated above with respect to claim 38.

Thus, the Examiner erred in rejecting claims 4, 8-10, 12, 17, 20-21, 27-28, 31, 36-37, 39, 41-42, and 50-51 under U.S.C. §103(a) based upon the Howard and Crandall patents. Reconsideration and reversal of the rejections of claims 4, 8-10, 12, 17, 20-21, 27-28, 31, 36-37, 39, 41-42, and 50-51 accompanied by allowance of claims 4, 8-10, 12, 17, 20-21, 27-28, 31, 36-37, 39, 41-42, and 50-51 are respectfully requested.

III. Conclusion.

For the foregoing reasons the Examiner erred in rejecting claims 1-51. Reconsideration and reversal of the rejections of claims 1-51 accompanied by allowance of claims 1-51 are respectfully requested.

Respectfully submitted,

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Appendix A
CLAIMS ON APPEAL (1-51)

1. (Original) An adhesive composition, the adhesive composition comprising:
a plurality of cationic microspheres; and
a fluidizing medium effective for supporting fluid application of the adhesive composition to a surface.
2. (Original) The adhesive composition of claim 1 wherein the cationic microspheres have a cationic charge that is permanently and individually bound to each cationic microsphere.
3. (Original) The adhesive composition of claim 1 wherein the cationic microspheres are derived in part from a cationic unsaturated vinyl monomer.
4. (Original) The adhesive composition of claim 1 wherein the cationic microspheres are derived in part from a CN alkyl(meth)acrylate monomer where N is any integer ranging from 4 to 14.
5. (Original) The adhesive composition of claim 1 wherein the cationic microspheres are derived in part from an unsaturated vinyl comonomer.
6. (Original) The adhesive composition of claim 1 wherein the adhesive composition further comprises a cationic latex adhesive binder.

Appendix A (Continued)
CLAIMS ON APPEAL (1-51)

7. (Original) The adhesive composition of claim 1 wherein the adhesive composition further comprises a cationic surfactant.
8. (Original) An adhesive article, the adhesive article comprising:
 - a substrate; and
 - a coating of the adhesive composition of claim 1 on the substrate, the adhesive composition effective to allow positioning of the adhesive article on a first application surface and repositioning of the adhesive article on a second application surface.
9. (Original) The adhesive article of claim 8 wherein the adhesive composition is effective to allow the repositioning of the adhesive article from the first application surface to the second application surface without leaving any visible residue of the pressure sensitive adhesive on the first application surface, the visible residue being any residue that is visible to the unaided eye of a human being.
10. (Original) A method of making an adhesive article, the method comprising applying a coating of the adhesive composition of claim 1 on a substrate to form the adhesive article, the adhesive composition effective to allow positioning of the adhesive article on a first application surface and repositioning of the adhesive article on different application surfaces.

Appendix A (Continued)
CLAIMS ON APPEAL (1-51)

11. (Original) A mixture, the mixture comprising polymerizable substances, the polymerizable substances comprising:

at least one CN alkyl(meth)acrylate monomer, where N is any integer ranging from 4 to 14; and
a cationic unsaturated vinyl comonomer.

12. (Original) The mixture of claim 11 wherein the at least one CN alkyl(meth)acrylate monomer comprises iso-octyl acrylate.

13. (Original) The mixture of claim 11 wherein the polymerizable substances further comprise an unsaturated vinyl comonomer.

14. (Previously Presented) A coating composition, the coating composition comprising:
cationic microspheres based on the mixture of claim 11; and wherein the mixture further comprises a cationic latex adhesive binder.

15. (Previously Presented) The coating composition the mixture of claim 14 wherein the mixture further comprises a cationic surfactant.

Appendix A (Continued)
CLAIMS ON APPEAL (1-51)

16. (Original) The mixture of claim 11 wherein the mixture further comprises a cationic surfactant.
17. (Original) The mixture of claim 11 wherein the mixture further comprises a catalyzation initiator.
18. (Previously Presented) The mixture of claim 11 wherein polymerization of the polymerizable substances is effective to form cationic microspheres.
19. (Original) An adhesive, the adhesive comprising:
 - a polymerized product of polymerizable substances, the polymerized product adhesively repositionable between different application surfaces, the polymerizable substances comprising:
 - at least one CN alkyl(meth)acrylate monomer, where N is any integer ranging from 4 to 14; and
 - a cationic unsaturated vinyl comonomer.
20. (Original) The adhesive of claim 19 wherein the polymerized product comprises polymeric elastomeric microspheres that render the adhesive pressure sensitive.

Appendix A (Continued)

CLAIMS ON APPEAL (1-51)

21. (Original) The adhesive of claim 19 wherein the at least one CN alkyl(meth)acrylate monomer comprises iso-octyl acrylate.
22. (Original) The adhesive of claim 19 wherein the polymerizable substances further comprise an unsaturated vinyl comonomer.
23. (Original) The adhesive of claim 19 wherein the repositionable pressure sensitive adhesive further comprises a cationic latex adhesive binder.
24. (Original) The adhesive of claim 19 wherein the repositionable pressure sensitive adhesive further comprises a cationic surfactant.
25. (Original) The adhesive of claim 19 wherein the adhesive, when subjected to static angle testing on a surface of a painted steel panel, following coating of the adhesive as an adhesive stripe about 18 mm wide and about 33 mm long on a paper substrate at the rate of about 0.65 grams of the adhesive per square foot (7.0 grams per square meter) of the adhesive stripe, detaches from the surface of the painted steel panel about 300 seconds after being adhered to the surface of the painted steel panel.

Appendix A (Continued)

CLAIMS ON APPEAL (1-51)

26. (Original) The adhesive of claim 23 wherein the adhesive, when subjected to static angle testing on a surface of a painted steel panel, following coating of the adhesive as an adhesive stripe about 18 mm wide and about 33 mm long on a paper substrate at the rate of about 0.55 grams of the adhesive per square foot (5.9 grams per square meter) of the adhesive stripe, detaches from the surface of the painted steel panel about 240 seconds after being adhered to the surface of the painted steel panel.

27. (Original) An adhesive article, the adhesive article comprising:

a substrate; and

a coating of the adhesive of claim 19 on the substrate, the adhesive effective to allow positioning of the adhesive article on a first application surface and repositioning of the adhesive article on a second application surface.

28. (Original) The adhesive article of claim 27 wherein the adhesive composition is effective to allow the repositioning of the adhesive article from the first application surface to the second application surface without leaving any visible residue of the pressure sensitive adhesive on the first application surface, the visible residue being any residue that is visible to the unaided eye of a human being.

Appendix A (Continued)

CLAIMS ON APPEAL (1-51)

29. (Original) A method of making an adhesive composition, the method comprising:
causing a plurality of cationic microspheres to exist in a fluidizing medium to
yield the adhesive composition, the fluidizing medium effective for
supporting fluid application of the adhesive composition to a surface.
30. (Original) The method of claim 29 wherein the cationic microspheres have a cationic charge
that is permanently and individually bound to each cationic microsphere.
31. (Original) The method of claim 29 wherein the cationic microspheres are derived in part
from a CN alkyl(meth)acrylate monomer where N is any integer ranging from 4 to 14.
32. (Original) The method of claim 29 wherein the cationic microspheres are derived in part
from an unsaturated vinyl comonomer.
33. (Original) The method of claim 29, the method further comprising incorporating a cationic
latex adhesive binder in the adhesive composition.

Appendix A (Continued)

CLAIMS ON APPEAL (1-51)

34. (Original) The method of claim 33, the method further comprising incorporating a cationic surfactant in the adhesive composition.

35. (Original) The method of claim 29, the method further comprising incorporating a cationic surfactant in the adhesive composition.

36. (Previously Presented) An adhesive article, the adhesive article comprising:

a substrate; and

a coating of the adhesive composition prepared in claim 29 on the substrate, the adhesive composition effective to allow positioning of the adhesive article on a first application surface and repositioning of the adhesive article on a second application surface.

37. (Previously Presented) The adhesive article of claim 36 wherein the adhesive composition is effective to allow the repositioning of the adhesive article from the first application surface to the second application surface without leaving any visible residue of the pressure sensitive adhesive composition on the first application surface, the visible residue being any residue that is visible to the unaided eye of a human being.

Appendix A (Continued)

CLAIMS ON APPEAL (1-51)

38. (Original) A method of making adhesive cationic microspheres, the method comprising:
initiating reaction of a polymerizable mixture to form the adhesive cationic
microspheres, the polymerizable mixture comprising:
at least one CN alkyl(meth)acrylate monomer, where N is any integer ranging
from 4 to 14; and
a cationic unsaturated vinyl comonomer.
39. (Original) The method of claim 38 wherein the at least one CN alkyl(meth)acrylate
monomer comprises iso-octyl acrylate.
40. (Original) The method of claim 38 wherein the polymerizable mixture further comprises an
unsaturated vinyl comonomer.
41. (Original) The method of claim 38 wherein initiating reaction of the polymerizable mixture
comprises:
including a catalyzation initiator in the polymerizable mixture; and
triggering catalyzation initiation by the catalyzation initiator.

Appendix A (Continued)

CLAIMS ON APPEAL (1-51)

42. (Original) The method of claim 41 wherein triggering catalyzation initiation by the catalyzation initiator comprises warming the catalyzation initiator to cause free radical generation by the catalyzation initiator.
43. (Original) The method of claim 38, the method further comprising including a cationic surfactant in the polymerizable mixture.
44. (Original) A method of using the adhesive cationic microspheres of claim 38, the method comprising uniformly mixing the adhesive cationic microspheres with a cationic latex adhesive binder to form an adhesive composition.
45. (Original) The method of claim 44, the method further comprising including a cationic surfactant in the polymerizable mixture.
46. (Original) The method of claim 45, wherein the adhesive composition, when subjected to static angle testing on a surface of a painted steel panel, following coating of the adhesive composition as an adhesive stripe about 18 mm wide and about 33 mm long on a paper substrate at the rate of about 0.65 grams of the adhesive composition per square foot (7.0 grams per square meter) of the adhesive stripe, detaches from the surface of the painted steel panel about 300 seconds after being adhered to the surface of the painted steel panel.

Appendix A (Continued)

CLAIMS ON APPEAL (1-51)

47. (Original) A method of using the adhesive cationic microspheres of claim 40, the method comprising uniformly mixing the adhesive cationic microspheres with a cationic latex adhesive binder to form an adhesive composition.

48. (Original) The method of claim 47, the method further comprising including a cationic surfactant in the polymerizable mixture.

49. (Original) The method of claim 48, wherein the adhesive composition, when subjected to static angle testing on a surface of a painted steel panel, following coating of the adhesive composition as an adhesive stripe about 18 mm wide and about 33 mm long on a paper substrate at the rate of about 0.55 grams of the adhesive composition per square foot (5.9 grams per square meter) of the adhesive stripe, detaches from the surface of the painted steel panel about 240 seconds after being adhered to the surface of the painted steel panel.

50. (Original) An adhesive article, the adhesive article comprising:
a substrate; and
a coating of the adhesive composition prepared in claim 44 on the substrate, the adhesive composition effective to allow positioning of the adhesive article on a first application surface and repositioning of the adhesive article on a second application surface.

Appendix A (Continued)

CLAIMS ON APPEAL (1-51)

51. (Original) The adhesive article of claim 50 wherein the adhesive composition is effective to allow the repositioning of the adhesive article from the first application surface to the second application surface without leaving any visible residue of the pressure sensitive adhesive on the first application surface, the visible residue being any residue that is visible to the unaided eye of a human being.

Appendix B
REFERENCES CITED BY THE EXAMINER
(Copies Are Attached)

- * U.S. Patent No. 4,598,112 (Howard)
- * U.S. Patent No. 5,756,625 (Crandall)

Inventor: Andrew J. Callinan

Serial No.:10/725,634

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Appendix C
RELATED PROCEEDINGS
(There are no related proceedings)

United States Patent [19]
Howard

[11] **Patent Number:** **4,598,112**
[45] **Date of Patent:** **Jul. 1, 1986**

[54] **LOW TACK CATIONIC MICROSPHERE
GLUE**

[75] **Inventor:** **Philip H. Howard, Jamesville, N.Y.**

[73] **Assignee:** **International Cube Corp., Syracuse,
N.Y.**

[21] **Appl. No.:** **650,448**

[22] **Filed:** **Sep. 14, 1984**

[51] **Int. Cl.⁴** **C08K 11/00**

[52] **U.S. Cl.** **524/78; 523/223;
524/460**

[58] **Field of Search** **524/78, 460; 523/223**

[56] **References Cited**

U.S. PATENT DOCUMENTS

3,954,967 5/1976 Urton 424/81

4,166,152 8/1979 Baker et al. 524/832
4,215,162 7/1980 Kunnen et al. 524/724
4,495,318 1/1985 Howard 524/460

Primary Examiner—Joseph L. Schofer.

Assistant Examiner—J. M. Reddick

Attorney, Agent, or Firm—Bruns and Wall

[57] **ABSTRACT**

Inherently tacky, elastomeric, solvent-dispersible, solvent-insoluble, polymeric microspheres prepared using a cationic emulsifier. The tacky microspheres are particularly adapted for use as a low tack microsphere glue that can be applied to a sheet material substrate whereby the latter is provided with a reusable adhesive surface.

1 Claim, No Drawings

LOW TACK CATIONIC MICROSPHERE GLUE

RELATED APPLICATION

This application is related as to subject matter to copending application Ser. No. 591,948, filed Mar. 21, 1984, now U.S. Pat. No. 4,495,318, by Philip H. Howard for Low Tack Microsphere Glue. Said copending application is owned by the assignee of the present invention.

BACKGROUND OF THE INVENTION

This invention relates generally to tacky polymeric microspheres, and has particular reference to inherently tacky, elastomeric, solvent-dispersible, solvent-insoluble, polymeric microspheres and a process for preparing same using a cationic emulsifier.

In U.S. Pat. No. 4,166,152, granted Aug. 28, 1979 to W. A. Baker et al., there is a disclosure of inherently tacky acrylate homopolymer microspheres prepared by aqueous suspension polymerization techniques utilizing a suspension stabilizer and an anionic emulsifier. The microspheres prepared by this method had an average particle size of 10-60 microns. The anionic emulsifiers retard particle coalescence by surrounding the particle with a negatively charged double layer which provides repulsion of the individual particles.

The inherently tacky, negatively charged microspheres disclosed by Baker have desirable properties in many applications where a low tack, reusable adhesive surface is desirable. All of these applications require that the microspheres be attached to sheet material as is disclosed for copolymer microspheres in U.S. Pat. No. 3,857,731, granted Dec. 31, 1974 to R. F. Merrill, Jr. et al. The Merrill et al. patent suggests that copolymer microspheres, which also were prepared with an anionic emulsifier, can be anchored to the sheet material using a film-forming binder in which the copolymer microspheres are embedded. For some applications, it is desirable to have microspheres that are cationically (positively) charged to facilitate attachment to the sheet material. For example, paper has a slight anionical charge which should bond strongly to cationically charge microspheres. Comparisons of anionic and cationic surfactant microspheres coated on paper from a hexane solution demonstrate that the cationic microspheres are tightly bonded to paper and do not have the transference problems frequently noted with the anionic microspheres.

Other pertinent prior patents of which the applicant is aware are U.S. Pat. Nos. 3,513,120; 3,691,140; 3,857,731; 4,049,483 and 4,049,604.

SUMMARY OF THE INVENTION

The present invention is directed to inherently tacky microspheres having similar properties to those disclosed by the Baker patent but which are prepared using a cationic emulsifier. The tacky microspheres provided by the invention are particularly adapted for use as a low tack microsphere glue that can be applied to a sheet material substrate whereby the latter is provided with a reusable adhesive surface especially on paper.

DESCRIPTION OF THE PREFERRED EMBODIMENT

In the present invention, the low tack microsphere glue is essentially comprised of inherently tacky, elastomeric,

solvent-dispersible, solvent-insoluble, polymeric microspheres (having an average particle size of 20-60 microns) prepared using a cationic emulsifier. The microspheres are derived from non-ionic monomers and included a water emulsifiable methacrylate ester, or alkyl acrylate. The microspheres are prepared by aqueous suspension polymerization techniques using a suspension stabilizer and, as noted, a cationic emulsifier.

In a preferred embodiment of the invention, the monomer is 2-ethylhexyl acrylate, the stabilizer is an aqueous solution of polyacrylic acid, and the cationic emulsifier is an ethoxylated tallow amine. A catalyst is also employed in the polymerization of the monomer, the catalyst being a benzoyl peroxide in the preferred embodiment.

EXAMPLE 1

To a 5 liter, 3-necked Morton flask equipped with thermometer, mechanical stirrer, reflux condenser, and a vacuum and nitrogen inlet tube, were added 2820 grams of deionized or distilled water and 54 grams of Carbopol EX-17 (trademark for a 15% active aqueous solution of polyacrylic acid of 300,000-500,000 molecular weight range commercially available from the B. F. Goodrich Co.). The contents of the flask were then mixed to dissolve the EX-17. Concentrated ammonium hydroxide was then added to the mixture until a pH of 7.0 was achieved.

To this solution were added 3.5 grams of Lucido-70 (trademark for a 70% active benzoyl peroxide in water; granular, bulk density—55 lbs./ft.³, freezing point less than 32° F.; commercially available from Lucidol Division, Penwalt Corp.), 1000 grams of 2-ethylhexyl acrylate, and 30 grams of Trymeen TAM-20 (trademark for an ethoxylated tallow amine, (20 average moles of ethylene oxide) cationic surfactant (liquid at 25° C., pour pt. —2° C., HLB=15.4, viscosity at 100° F.=119 cSt), commercially available from Emery Industries, Inc.). Vacuum was then drawn upon the contents of the flask, the pressure therein being drawn to approximately 25 inches of mercury, and held for approximately fifteen minutes (while the mixture is stirred via vacuum tight stir assembly) to assure removal of dissolved air and oxygen. The vacuum was then broken with nitrogen. A nitrogen purge was maintained throughout the emulsion and polymerization steps. Agitation for the mixture was set at 300 r.p.m.

The batch was then heated to 60° C. and maintained for 16 hours. As the temperature initially approached 60° C., a mild exotherm was noted which raised the temperature to approximately 70° C. After 16 hour period, the suspension was passed through a 250 micron screen. The resultant homopolymer aqueous emulsion contained approximately 26.3% solids. Upon standing, the polymer spheres creamed to the surface, but were readily dispersed by agitating the mixture. Particle size ranged from 15-60 microns, with the average size approximately 35 microns.

Average tack level for the microsphere glue is 98 grams of tack as determined by a Polyken, Jr. Probe Track tester. The Polyken, Jr. tester is manufactured by Testing Machines, Inc., under license from the Kendall Corp., and coating was accomplished by drawdown technique using a No. 20 wirewound rod. Values represent averages obtained from several testings of the coated paper. Conditions of testing are as follows:

Contact time: 1 second

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Separation time: 1 cm/sec

Contact pressure: 100 g/cm²

Probe type: $\frac{1}{8}$ cm diameter 304 stainless steel tip—280 grit abrasive finish

Temperature: approximately 22.5° C.

Further information on the Polyken probe tack tester may be found in ASTM D29 79 Section 15.06, 1983.

EXAMPLE 2

Aqueous microspheres of the preferred embodiment as well as microspheres synthesized with an anionic emulsifier Siponate DS-10 (sodium dodecyl benzene sulfonate, commercially available from Alcolac, Inc.) as described by Baker et al, U.S. Pat. No. 4,166,152, were each coagulated with methanol, and then rinsed with methanol to drive off the water. Samples of each were placed in a forced air oven at 150° F. for 30 minutes and then placed in a desiccator to cool. Fifteen grams of the dried spheres were then placed in respective 600 ml beakers to which was added 200 ml of hexane. The samples were stirred with a magnetic stir bar until the spheres were fully redispersed in the solvent.

Samples of each dispersion were then coated onto unprimed and untreated 50 lb. James River Mill paper with a No. 36 wire wound rod to produce coating weights which ranged from 1.5–2.5 lbs./300 sq. ft. Samples were dried in a forced air oven for 5 minutes at 150° F. and then cooled on the lab bench for approximately 30 minutes.

Samples measuring 11.25 sq. in. were cut from the coated papers and pressed by hand onto both Hammermill Xerox paper and Westvaco 50 lb. Clear Spring paper. Samples were pressed with both light and firm pressure by hand. In all cases, the microspheres synthesized with the cationic emulsifier showed no sign of transfer to the uncoated test papers, whereas those microspheres prepared with the anionic emulsifier transferred to a significant degree in each test. The non-ionic microspheres transferred in an increasing amount

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with heavier pressure, whereas the cationic microspheres did not transfer with even the heaviest pressure exerted by hand.

From the foregoing description it will be apparent that the invention provides an improved low tack microsphere glue having advantages over the prior art. As will be understood by those familiar with the art, the invention may be embodied in other specific forms without departing from the spirit or essential characteristics thereof.

I claim :

1. A low tack microsphere containing glue suspension for application to a substrate to provide a reusable adhesive surface, wherein the microspheres are surrounded by a cationic surfactant which prevents particle coalescence, prepared according to the process comprising the steps of:

admixing, in approximate parts by weight, 2820 parts water; 54 parts of a 15% aqueous solution of polyacrylic acid having a molecular weight of 300,000 to 500,000 until said acid is dissolved, to form a solution;

raising the pH of said solution to 7.0 by adding concentrated ammonium hydroxide thereto;

adding 3.5 parts of a 70% active benzoyl peroxide catalyst, 1000 parts 2-ethylhexyl acrylate and 30 parts of an ethoxylated tallow amine to said solution to form a mixture;

holding said mixture under a vacuum of about 25 inches of mercury with stirring for about 15 minutes;

heating said mixture to about 60° C. for about 16 hours to form an aqueous suspension; and

passing said suspension through a 250 micron screen to remove a small amount of unsuitable larger particle agglomerates.

* * * * *



US005756625A

United States Patent [19]

Crandall et al.

[11] Patent Number: 5,756,625

[45] Date of Patent: May 26, 1998

[54] STABILIZED ADHESIVE MICROSPHERES

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[22] Filed: Oct. 11, 1996

[51] Int. Cl.⁶ C08F 18/02; C08F 218/02;
C08F 20/10; C08F 12/30[52] U.S. Cl. 526/320; 523/218; 523/223;
526/278; 526/287; 526/307; 526/307.7;
526/312[58] Field of Search 526/278, 287,
526/307, 307.7, 312, 320; 523/223, 218

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U.S. PATENT DOCUMENTS

3,620,988	11/1971	Cohen	524/723
3,691,140	9/1972	Silver	526/240
3,857,731	12/1974	Memill, Jr. et al.	428/314.4
4,166,152	8/1979	Baker et al.	428/522
4,276,212	6/1981	Khanna et al.	524/512
4,495,318	1/1985	Howard	524/375
4,598,112	7/1986	Howard	524/78
4,645,783	2/1987	Kinoshita	523/221
4,656,218	4/1987	Kinoshita	524/460
4,735,837	4/1988	Miyasaka et al.	428/40
4,786,696	11/1988	Bohnel	526/88
4,952,650	8/1990	Young et al.	526/194
4,988,567	1/1991	Delgado	428/402
5,053,436	10/1991	Delgado	521/64

5,215,818	6/1993	Silver et al.	428/343
5,571,617	11/1996	Coopridge et al.	428/341

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[57] ABSTRACT

Stabilized microsphere adhesive composition comprising: a plurality of polymeric, elastomeric microspheres wherein the microspheres are the reaction product of reactants comprising polymerizable starting materials comprising at least one C₄–C₁₄ alkyl (meth)acrylate monomer and optionally at least one comonomer, at least one vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms in an amount about 0.1 to 3 parts by weight of the microspheres; optionally, a polymeric stabilizer in an amount of between about 0.1 and about 3 parts by weight per 100 parts by weight of the microspheres; a surfactant in an amount of no greater than about 5 parts by weight per 100 parts by weight of the microspheres; optionally a modifier, wherein the modifier is at least one of a chain transfer agent, a tackifier, a solvent or the like in an amount that is sufficient to provide microspheres with a n-heptane soluble portion in the range of 30–98%; and (f) an initiator in an amount effective to catalyze the polymerization reaction present in amounts ranging from about 0.1 to approximately 2 parts by weight per 100 parts by weight of the polymerizable monomer starting material.

20 Claims, No Drawings

STABILIZED ADHESIVE MICROSPHERES

TECHNICAL FIELD

This invention relates to a composition of suspension polymerized adhesive microspheres and in particular to adhesive microspheres that exhibits enhanced adhesion to substrates while remaining repositionable and are more stable during polymerization and processing.

BACKGROUND OF THE INVENTION

Inherently tacky, elastomeric microspheres are known in the art to be useful in repositionable pressure-sensitive adhesive applications. As used herein, the term "repositionable" refers to the ability to be repeatedly adhered to and removed from a substrate without substantial loss of adhesion capability. Repositionable adhesives are used in the construction of temporary messaging or signage for the home and office environment. An example of such products are Post-it® self-stick notes sold by 3M Company.

Numerous references concern the preparation and/or use of inherently tacky, elastomeric acrylate polymeric microspheres which are solid in nature. Such spheres and their use in aerosol adhesive systems having repositionable properties are disclosed in U.S. Pat. No. 5,215,818. These microspheres are prepared by aqueous suspension polymerization of alkyl acrylate monomers and ionic comonomers, for example, sodium methacrylate, in the presence of emulsifier-preferably an anionic emulsifier. The ionic comonomers other than a mentioned are specifically zwitterionic in nature and are not shown to enhance suspension stability or performance.

U.S. Pat. No. 4,166,152 describes solid, inherently tacky (meth)acrylate microspheres which are prepared from non-ionic alkyl acrylate or methacrylate monomer(s) in the presence of both an emulsifier and an ionic suspension stabilizer having an interfacial tension sufficient to prevent microsphere agglomeration. Such microspheres are also disclosed in U.S. Pat. Nos. 4,495,318 and 4,598,112, where the preparative methods involve the use of a non-ionic emulsifier or a cationic emulsifier. All three patents disclose utility as a "reusable adhesive".

Increasing the adhesion of these types of adhesives has been tried by various investigators: U. S. Pat. No. 5,053,436 provides a hollow core in the microsphere. In this morphology the microsphere is said to offer increased resistance to adhesive transfer and an increased adhesion level due to the hollow nature of the microsphere. U. S. Pat. No. 4,988,567 describes microspheres having multiple small voids. These voids are said to enhance the adhesive properties of the microsphere. The microspheres described are said to be solvent insoluble.

U.S. Pat. No. 4,786,696 describes a suspension polymerization process for preparing solid, inherently tacky (meth)acrylate microspheres which does not require the use of either an ionic comonomer or an ionic suspension stabilizer in order to prevent agglomeration. Rather, the process requires agitation of the vessel charge prior to the initiation of the reaction sufficient to create a suspension of monomer droplets having an average monomer droplet size of between about 5 and about 70 micrometers. In addition to (meth)acrylate monomer, a minor portion of a non-ionic, vinylic comonomer such as e.g., acrylic acid may be included to modify the "tacky nature" of the microspheres. The resultant adhesive microspheres exhibit high tack values thought to be due to the lack of stabilizer on the microsphere surface.

U.S. Pat. No. 3,620,988 discloses a method of preparing "bead-type polymers" which involves the use of a water-

insoluble polymeric thickening dispersing agent. The method can be applied to produce pressure-sensitive adhesives in the form of coatable bead suspensions, the adhesives comprising a high solids suspension/dispersion of a lightly crosslinked polymer of a higher alkyl acrylate and a tackifier.

U.S. Pat. No. 4,735,837 discloses a detachable adhesive sheet having an adhesive layer containing "elastic microballs", wherein the microballs partially protrude from the surface of the adhesive layer. The microballs may or may not be tacky. They can be derived from, e.g., (meth)acrylate monomer and an α -olefinic carboxylic acid monomer via suspension polymerization in an aqueous medium. However, no details as to the nature of the surfactants utilized, etc., are disclosed. The microballs and an adhesive are dispersed in solvent, mixed, and coated, with the ratio of adhesive to microballs being from about 1:10 to about 10:1. This ratio is disclosed to be critical in order that all microballs in the final product, including those protruding from the surface, are completely covered with the adhesive. A range of 1,000 to 150,000 pieces per square centimeter is disclosed as preferred.

DE 3,544,882 A1 describes crosslinked microspheres composed of 90 to 99.5 weight percent of (meth)acrylate ester and 10 to 0.5 weight percent of vinyl type monomer, e.g., acrylic acid, having a reactive functional group through which crosslinking is achieved by reaction with an oil-soluble crosslinking agent. The microspheres are prepared by dispersing in water a solution (in organic solvent) of copolymer prepared by known methods such as solution, bulk, emulsion, or suspension polymerization. (However, the reference notes that in cases where emulsion or suspension polymerization is used with water as a dispersion medium, it is not necessary to make a new aqueous dispersion.) When tacky, the spheres are said to be useful in spray or coated sheet form as "removable adhesive". The stated purpose of the invention is to provide microspheres having a uniform particle size, but it is also stated that the microspheres may contain other monomers such as vinyl acetate, styrene, acrylonitrile, methacrylonitrile, etc., "... to prevent partial transfer of the adhesive when the carrier is pulled away from the substrate ...".

U.S. Pat. Nos. 4,645,783 and 4,656,218 disclose a "repeatably usable and releasable sheet" coated with an aqueous suspension of microspheres obtained by aqueous suspension polymerization (in the presence of a protective colloid comprising casein as a main ingredient) of one or more alkyl (meth)acrylate esters, α -monoolefin carboxylic acids, and one or more other vinyl monomers.

The microspheres are preferably interspersed with finer polymer particles prepared by polymerization of one or more vinyl monomers in an aqueous medium. These fine polymeric particles are said to be "... effective in improving the anchorage to the adherend and the adhesion to the substrate after the aqueous suspension prepared in accordance with the present innovation is applied to the substrate".

U.S. Pat. No. 3,857,731 and EP 209,337 both address problems with adhesive microspheres transfer. The former discloses sheets coated with the tacky elastomeric copolymer microspheres of the Silver patent (U.S. Pat. No. 3,691,140) and a binder material which provides sockets in which the microspheres are held by predominantly mechanical forces. The latter states that adhesive microspheres could be put to more demanding applications if it were not for the drawback of adhesive transfer. Tacky, elastomeric micro-

spheres are then described which have a composition formed from non-ionic monomers alone or together with a proportion of ionic comonomers wherein the ionic monomers are first dissolved in an organic cosolvent. The microspheres further comprise an adhesion-promoting monomer having functionality which remains unreacted during polymerization of the monomers and is available for subsequently binding the microspheres through electrostatic interaction or chemical bonding to a substrate or binder-coated substrate. Preferably, the microspheres are derived from at least one alkyl acrylate or methacrylate ester.

U.S. Pat. No. 5,326,842 describes the use of a dual polymerization process comprising first a suspension polymerization process, followed by an emulsion polymerization. This dual polymerization allows for low or high tack coatings to be prepared. For high tack coatings a chain transfer agent is used in the suspension step, the subsequent emulsion polymerization provides the stability of the final material.

In U.S. Pat. No. 4,952,650, the polymerization of large size, nontacky beads that are subsequently extruded into permanent film adhesives is described. In this disclosure styrene sulfonic acid or the sodium salt thereof is in a general listing of monomers, however it is not used in the examples or said to provide novel properties of the material.

Other investigators have prepared polyacrylate beads for use in microporous sorbents as binding agents. A polyacrylate polymer product comprising (a) a suspension stabilizer modifier having a metal cation and (b) a copolymer of acrylic acid ester of a non-tertiary alcohol having 1 to 14 carbon atoms, a polar monomer, and at least one of the following: a higher vinyl ester, styrene sulfonate salt, multi-vinyl monomer, and α , β -ethylenically unsaturated poly(alkyleneoxy) is exemplified. The use of suspension stabilizers such as zinc oxide and colloidal silica, as described, are known to suppress the adhesion characteristics of the resulting polymer. In addition very high levels (approximately 5.5% by weight to monomer) of the surfactant and styrene sulfonate salt are used. Again such excessive levels are detrimental to adhesion characteristics and would limit the utility of these polymeric beads as repositionable adhesives.

It has now been discovered that microspheres that contain vinyl unsaturated additives with both an ionic and hydrophobic moiety to enhance stability and performance properties, while maintaining inherent tackiness, elastomeric properties and solvent or water dispersibility.

SUMMARY OF THE INVENTION

Advantageously, the present invention provides a microsphere adhesive that enhances adhesion by avoiding microsphere surface contaminants, without changing the physical morphology of the microsphere, or by co-polymerizing a secondary polymer reaction. Uniquely, the microspheres of the present invention are prepared with the use of specialty ionic monomers that serve not only as a stabilizer but enhance the performance properties of the resulting adhesive.

This invention provides pressure sensitive adhesive microspheres that contain vinyl unsaturated additives with both an ionic and hydrophobic moiety to enhance stability and performance properties. Furthermore, these adhesive microspheres adhere to substrates, remove cleanly and are able to be reapplied multiple times if desired. Even with enhanced adhesion the adhesive microspheres of the present invention adhere non-destructively to fragile surfaces such as paper.

In particular, the present invention provides an adhesive microsphere composition comprising:

- (a) a plurality of polymeric, elastomeric microspheres wherein the microspheres are the reaction product of reactants comprising polymerizable starting materials comprising at least one C_4 - C_{14} alkyl (meth)acrylate monomer and optionally at least one comonomer;
- (b) from 0.1 to 3 parts by weight of at least one vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms, preferably about 0.5 to about 3 parts by weight of the microspheres;
- (c) optionally, a polymeric stabilizer in an amount of between about 0.1 and about 3 parts by weight per 100 parts by weight of the microspheres, preferably about 0.1 to about 1.5 parts by weight per 100 parts by weight of the microspheres;
- (d) a surfactant in an amount of no greater than about 5 parts by weight per 100 parts by weight of the microspheres, preferably no greater than 3 parts by weight and most preferably in the range of 0.2 to about 1.5 parts by weight per 100 parts by weight of the microspheres;
- (e) optionally a modifier, wherein the modifier can be at least one of a chain transfer agent, a tackifier, a solvent or the like in an amount that is sufficient to provide microspheres with a solvent soluble portion in the range of 30-98%, preferably in the range of 40-95%, wherein the solvent is n-heptane; and
- (f) an initiator in an amount effective to catalyze the polymerization reaction present in amounts ranging from about 0.1 to approximately 2 parts by weight per 100 parts by weight of the polymerizable monomer starting material.

As used in this application, the notation "(meth)acrylate" refers to acrylate and methacrylate.

The vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms improves the stability of the suspension polymerization process resulting in little coagulation when compared to the formulations not containing it. In addition the use of the vinyl-unsaturated additive results in enhanced adhesive performance. Furthermore high solids content, stable suspension polymerizations can now be made.

A modifier may be used to regulate the solvent soluble portion of the microspheres and it is added to the polymerization mixture in an amount sufficient to provide a solvent soluble portion that is in the range of 30-98%, preferably in the range of 40-95%. Various modifiers may be used within the scope of this invention and the amounts used are those that sufficiently provide the microspheres with a solvent soluble portion. Such amounts would range, for example for solvents from 1-30%, for tackifiers from 1-30% and for chain transfer agents, up to about 0.15%.

Particularly useful modifiers are chain transfer agents. To control the molecular weight of the polymer being formed in the microsphere a chain transfer agent or modifier is used. Many halogen- and sulfur-containing organic compounds function well as chain transfer agents in free radical polymerizations. Non-limiting examples of such agents are: carbon tetrabromide, carbon tetrachloride, dodecanethiol, iso-octylthioglycolate, butyl mercaptan, and tertiary-dodecyl mercaptan. Particularly useful chain transfer agents are long chain mercaptans, such as dodecanethiol. The amount of chain transfer agent suitable for microsphere

polymerizations is calculated on a weight basis to the entire polymerizable content. The chain transfer agent is preferably added at up to about 0.15%, more preferably up to about 0.12% and most preferably up to about 0.08%. These levels are adequate to provide a solvent soluble polymer component in the microsphere of up to about 98%.

Other useful modifiers are solvents. Examples of which are but not limited to: aliphatic or aromatic solvents such as heptane, benzene, toluene and the like; alcohols such as methanol, isopropyl alcohol, and the like; and ketones such as acetone, methyl ethyl ketone and the like. The amount of solvent suitable for microsphere polymerizations is calculated on a weight basis to the entire polymerizable content. The solvent is preferably added at up to about 30%, more preferably up to about 15% and most preferably up to about 5%. These levels are adequate to provide a solvent soluble polymer component in the microspheres of up to about 98%.

Still other useful modifiers include tackifiers and/or plasticizers. Examples of which are but not limited to: hydrogenated rosin esters commercially available from such companies as Hercules, Inc. under the tradenames of Foral™, Regalrez® and Pentalyn™. Tackifying resins also include those based on t-butyl styrene. Useful plasticizers include but are not limited to dioctylphthalate, 2-ethylhexyl phosphate, tricresyl phosphate, mineral oil and the like. The tackifier and/or plasticizer are preferably added at up to about 30%, more preferably up to about 15% and most preferably up to about 5%. These levels provide a solvent soluble polymer component in the microsphere of up to about 98%.

In another aspect of the present invention a one step suspension polymerization process is provided for preparing polymeric elastomeric microspheres comprising the steps of:

- (a) stirring or agitating a mixture comprising polymerizable monomer starting materials comprising:
 - (i) at least one C_4-C_{14} alkyl (meth)acrylate monomer and, optionally at least one comonomer;
 - (ii) from 0.1 to 5 parts by weight of at least one vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms;
 - (iii) an initiator for the polymerizable monomer starting materials present in amounts ranging from 0.1 to approximately 2 parts per weight per 100 parts by weight of the polymerizable monomer starting materials;
 - (iv) optionally, a polymeric stabilizer in an amount in the range of 0.1 to about 3 parts by weight per 100 parts by weight of the polymerizable monomer starting materials;
 - (v) a surfactant in an amount no greater than about 5 parts by weight per 100 parts by weight of polymerizable monomer, preferably no greater than about 3 parts by weight and most preferably in the range of 0.5 to 1.5 parts by weight;
 - (vi) water to form an oil in water suspension; and
 - (vii) optionally a modifier in an amount sufficient to provide a solvent soluble portion in the range of 30-98%; and
- (b) polymerizing the (meth)acrylate monomer(s) and the comonomer(s), if present; wherein microspheres are provided.

In yet another aspect, the present invention provides a two-step suspension polymerization process for preparing polymeric elastomeric microspheres from polymerizable monomer starting materials, wherein the process comprising the steps of:

- (a) stirring or agitating a mixture comprising:
 - (i) at least one C_4-C_{14} alkyl (meth)acrylate monomer;
 - (ii) from 0.1 to 5 parts by weight of at least one vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms;
 - (iii) an initiator for the monomer present in amounts ranging from 0.1 to approximately 2 parts per weight per 100 parts by weight of the polymerizable monomer starting materials;
 - (iv) optionally, a polymeric stabilizer in an amount in the range of 0.1 to about 3 parts by weight per 100 parts by weight of the polymerizable monomer starting materials;
 - (v) a surfactant in an amount of no greater than about 5 parts by weight per 100 parts by weight of the polymerizable monomer starting materials, preferably no greater than 3 parts by weight and most preferably in the range of 0.5 to 2 parts by weight;
 - (vi) optionally a modifier in an amount sufficient to provide a solvent soluble portion in the range of 30-98%; and
 - (vii) water to form an oil in water suspension;
- (b) at least partially polymerizing the polymerizable monomer starting materials;
- (c) adding to the suspension at least one comonomer; and
- (d) continuing the polymerization of the polymerizable monomer starting materials; wherein microspheres are provided.

The present invention also provides in another aspect a sheet material comprising a backing and a coating of repositionable pressure sensitive adhesive described above is coated on at least one portion of at least one major surface.

Several features of the adhesive of the present invention provide a number of desirable advantages that have heretofore been unavailable. For example several advantages include, (a) improved adhesion to various surfaces (bond paper, fabric, wood, painted surfaces, glass, vinyl, etc), (b) adhesive microspheres which can be prepared at high solids (55% or greater) without large amounts of coagulum, (c) adhesive microspheres which are more stable to coagulation or agglomeration, and (d) adhesive microspheres that adhere to a substrate or backing and easily removes from applied surfaces without transferring or leaving an adhesive residue on the applied surface.

Further, the present invention provides a microsphere-based pressure sensitive adhesive having a high solvent soluble fraction that adheres to rough surfaces such as fabric, removes cleanly, and exhibits the ability to be reapplied multiple times if desired. Even with this enhanced adhesion to rough surfaces the adhesive microspheres will still adhere non-destructively to fragile surfaces such as paper. Furthermore, the adhesive microspheres of this invention is prepared according to resource efficient methods.

DESCRIPTION OF THE PREFERRED EMBODIMENTS(s)

The microspheres obtained in the present invention are the reaction product of (a) at least one alkyl (meth)acrylate ester wherein the alkyl group contains four to about 14 carbon atoms, preferably four to about 10 carbon atoms, (b) optionally, a comonomer, (c) at least one vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms, and (d) optionally, a modifier in an amount sufficient to provide a solvent soluble portion in the range of 30-98%.

The comonomer (b), if present may be nonpolar, ionic, polar or mixtures of such monomers.

Useful alkyl (meth)acrylate monomers are those monofunctional unsaturated (meth)acrylate esters, the alkyl groups of which have from 4 to 14 carbon atoms. Such (meth)acrylates are oleophilic, water dispersible, and are essentially water insoluble. Furthermore, useful (meth)acrylates are those that as homopolymers, generally have a glass transition temperature below about -20°C ., or if a combination of monomers is used, such a combination would produce a copolymer or terpolymer generally having a glass transition temperature below about -20°C . Nonlimiting examples of such (meth)acrylates included but are not limited to, isooctyl acrylate, 4-methyl-2-pentyl acrylate, 2-methylbutyl acrylate, isoamyl acrylate, sec-butyl acrylate, n-butyl acrylate, 2-ethylhexyl acrylate, isodecyl methacrylate, t-butyl acrylate, t-butyl methacrylate, isobornyl acrylate, methylmethacrylate, isononyl acrylate, isodecyl acrylate and the like, and the combination thereof.

Preferred alkyl (meth)acrylate monomers include isooctyl acrylate, isononyl acrylate, isoamyl acrylate, isodecyl acrylate, 2-ethylhexyl acrylate, n-butyl acrylate, sec-butyl acrylate and mixtures thereof.

Vinyl ester monomers suitable for use in the present invention include but are not limited to: vinyl 2-ethylhexanoate, vinyl capriate, vinyl laurate, vinyl pelargonate, vinyl hexanoate, vinyl propionate, vinyl decanoate, vinyl actanoate, and other monofunctional unsaturated vinyl esters of linear or branched carboxylic acids comprising 1 to 14 carbon atoms, which as homopolymers have glass transition temperatures below about -10°C . Preferred vinyl ester monomers include vinyl laurate, vinyl capriate, vinyl 2-ethylhexanoate, and mixtures thereof.

Additional other vinyl monomers which, as homopolymers, have glass transition temperatures higher than about -10°C . to 0°C ., such as vinyl acetate, acrylonitrile, styrene, mixtures thereof and the like, may optionally be utilized in conjunction with one or more of the acrylate, methacrylate and vinyl ester monomers provided the glass transition temperature of the resultant polymer is below about -10°C .

Suitable vinyl-unsaturated additives having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms include: the salts of sulfoesters of alpha-methylene carboxylic acids such as 2-sulfoethyl acrylate, 2-sulfoethyl methacrylate, 2-sulfoethyl alpha-ethylacrylate, 2-sulfoethyl alpha-hexylacrylate, 2-sulfoethyl alpha-cyclohexylacrylate, 2-sulfoethyl alpha-chloroacrylate, 2-sulfo-1-propyl acrylate, 2-sulfo-1-butyl acrylate and methacrylate, 3-sulfo-2-butyl acrylate and methacrylate, 2-methyl-1-sulfo-2-propyl acrylate and methacrylate, 3-bromo-2-sulfo-1-propyl acrylate, 3-chloro-2-sulfo-1-propyl acrylate, 3-chloro-2-sulfo-1-butyl acrylate, 3-methoxy-2-sulfo-1-propyl acrylate, 2-sulfo-cyclohexyl acrylate, 2-phenyl-2-sulfoethyl acrylate, 4-sulfo-1-butyl acrylate, 6-(sulfophenoxy) hexyl acrylate and methacrylate. The preparation of such materials is disclosed in U.S. Pat. No. 3,024,221, the disclosure of which is incorporated herein by reference.

The salts of sulfato-esters of alpha-methylene carboxylic acids including 3-sulfo-2-hydroxy-1-propyl methacrylate.

The salts of carboxy-terminated alkyl esters of alpha-methylene carboxylic acids including 11-methacryloxyundecanoic acid.

The salts of sulfoalkyl allyl ethers including 3-sulfo-2-hydroxy-1-propyl allyl ether.

The salts of acrylamide alkane sulfonates including 2-acrylamido-2-methyl propane sulfonates.

The salts of vinyl alkyl phosphonate esters including vinyl octyl phosphonates.

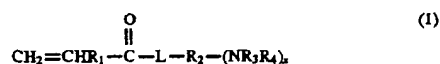
The salts of vinyl aryl sulfonates including para-styrene sulfonates.

Typically the vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety is present in relative amounts by weight of the total polymerizable content from about 0.1 to about 3 parts and preferably from about 0.5 to about 3 parts.

Suitable comonomers include nonpolar, ionic, polar monomers and mixtures thereof. In addition to using one or more acrylate monomers as a comonomer, as described above, the following are non-limiting examples of comonomers:

- (A) ionic comonomers, such as sodium methacrylate, ammonium acrylate, sodium acrylate, (I) trimethylamine p-vinyl benzimide, (II) 4,4, 9-trimethyl-4-azonia-7-oxo-8-oxa-dec-9-ene-1-sulphonate, (III) N,N-dimethyl-N-(beta-methacryloxyethyl) ammonium propionate betaine, (IV) trimethylamine methacrylimide, (V) 1,1-dimethyl-1(2,3-dihydroxypropyl)amine methacrylimide; any zwitterionic monomer and the like;
- (B) non-polar comonomers include but are not limited to, 4-methyl-2-pentyl acrylate, 2-methylbutyl acrylate, isoamyl acrylate, sec-butyl acrylate, n-butyl acrylate, isodecyl methacrylate, t-butyl acrylate, t-butyl methacrylate, isobornyl acrylate, octyl acrylamide, methylmethacrylate, isononyl acrylate, isodecyl acrylate, styrene and the like, and the combination thereof.
- (C) polar comonomers may or may not contain a dissociable hydrogen. Examples of suitable polar comonomers include organic carboxylic acids comprising 3 to about 12 carbon atoms and having generally 1 to about 4 carboxylic acid moieties. Nonlimiting examples of such monomers acrylic acid, methacrylic acid, itaconic acid, fumaric acid, crotonic acid, maleic acid, beta-carboxyethylacrylate and the like. In addition suitable polar comonomers include acrylamide, methacrylamide, 2-hydroxyethyl acrylate, and the like.

In addition, one class of suitable comonomers are amino-functional monomers having a nucleus or portion of the nucleus of the general formula (a):

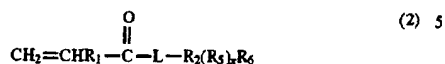


wherein

- R_1 is $-\text{H}$ -, $-\text{CH}_3$ -, $-\text{CH}_2\text{CH}_3$ -, cyano or carboxymethyl;
- R_2 is a hydrocarbyl radical comprising 1 to about 12 carbon atoms;
- R_3 and R_4 are independently H or an alkyl group containing 1 to about 12 carbon atoms or an arylalkyl group or together form a cyclic or heterocyclic moiety;
- L is carbon-carbon bond, O, NH or S; and
- x is an integer of 1 to 3.

Nonlimiting examples of comonomers according to formula (1) include N, N-dimethyl-aminoethyl(methyl)acrylate, N,N-dimethylaminopropyl-(meth)acrylate, t-butylaminoethyl(methyl)acrylate and N,N-diethylaminoacrylate.

Another class of suitable comonomers are comonomers having a nucleus or portion of the nucleus of the general formula (2):



wherein

R_1 is H , —CH_3 , $\text{—CH}_2\text{CH}_3$, cyano or carboxymethyl;

R_2 is a hydrocarbyl radical comprising 1 to about 12 carbon atoms;

R_3 is —O— , alkylene oxide comprising 1 to 5 carbon atoms, or phenoxy oxide, wherein the alkylene oxide would include, $\text{—CH}_2\text{O—}$, $\text{—CH}_2\text{CH}_2\text{O—}$, $\text{—CH}_2(\text{CH})\text{CH}_2\text{O—}$, and the like;

R_6 is H , $\text{—C}_6\text{H}_4\text{OH}$, or —CH_3 ;

L is a carbon—carbon bond, O , NH or S ; and

x is an integer with the proviso that when R_3 is —O— , x is an integer of 1–3.

Nonlimiting examples of comonomers according to formula (2) include hydroxyethyl (meth)acrylate, glycerol mono(meth)acrylate and 4-hydroxybutyl (meth)acrylate, acrylate terminated poly(ethylene oxide); methacrylate terminated poly(ethylene oxide); methoxy poly(ethylene oxide) methacrylate; butoxy poly(ethylene oxide) methacrylate; acrylate terminated poly(ethylene glycol); methacrylate terminated poly(ethylene glycol); methoxy poly(ethylene glycol) methacrylate; butoxy poly(ethylene glycol) methacrylate and mixtures thereof.

Yet another class of suitable comonomers are amido-functional monomers having a nucleus or portion of the nucleus of the general formula (3):



wherein

R_1 is H , —CH_3 , $\text{—CH}_2\text{CH}_3$, cyano or carboxymethyl; and

R_3 , R_4 are independently H or an alkyl group containing 1 to about 12 carbons or an arylalkyl group or together form a cyclic or heterocyclic moiety.

Nonlimiting examples of comonomers according to formula (3) include N -vinyl pyrrolidone, N -vinyl caprolactam, acrylamide or N , N -dimethyl acrylamide.

Nonlimiting examples of other suitable comonomers that do not fall within the above classes but are within the scope of permissible comonomers include (meth)acrylonitrile, furfuryl (meth)acrylate and tetrahydrofurfuryl (meth)acrylate, 2-vinyl pyridine, and 4-vinyl pyridine.

Typically, when a comonomer is present, the relative amounts by weight of the alkyl (meth)acrylate monomer(s) and the comonomer is in the range of about 99.5/0.5 to 75/25, and preferably is in the range of 98/2 to 92/8.

A modifier may be used to regulate the solvent soluble portion of the microspheres and it is added to the polymerization mixture in an amount sufficient to provide a solvent soluble portion that is in the range of 30–98%, preferably in the range of 40–95%. Various modifiers may be used within the scope of this invention and the amounts used are those that sufficiently provide the microspheres with a solvent soluble portion. Such amounts would range, for example for solvents from 5–30%, for tackifiers and/or plasticizers from 1–30% and for chain transfer agents, up to about 0.15%.

Particularly useful modifiers are chain transfer agents. To control the molecular weight of the polymer being formed in the microsphere it is desirable to use a chain transfer agent

or modifier. Many halogen- and sulfur-containing organic compounds function well as chain transfer agents in free radical polymerizations. Non-limiting examples of such agents are: carbon tetrabromide, carbon tetrachloride, dodecanethiol, iso-octylthioglycolate, butyl mercaptan, and tertiary-dodecyl mercaptan. In this invention it is efficacious to employ long chain mercaptans such as dodecanethiol. The amount of chain transfer agent suitable for these microsphere polymerizations is calculated on a weight basis to the entire polymerizable content. The chain transfer agent is preferably added at up to about 0.15% more preferably up to about 0.12% and most preferably up to about 0.08%. These levels are adequate to provide a soluble polymer content in the microsphere of up to about 98%.

The adhesive microspheres composition may also contain a crosslinking agent. Examples of useful crosslinking agents include, but are not limited to: multifunctional (meth)acrylate(s), e.g., butanediol diacrylate or hexanediol diacrylate or other multifunctional crosslinkers such as divinylbenzene and mixtures thereof. When used, crosslinker(s) is (are) added at a level of up to about 0.15 equivalent weight percent, preferably up to about 0.1 equivalent weight percent, of the total polymerizable composition with the proviso that the combination of crosslinking agent and modifier concentrations are chosen to obtain a microsphere with 30 to 98% solvent soluble portion.

The adhesive microspheres of the present invention are prepared by suspension polymerization using either a one-step or two-step process as described in detail below. Suspension polymerization is a procedure wherein a monomer is dispersed in a medium (usually aqueous) in which it is insoluble. The polymerization proceeds within the individual polymer droplets. Monomer soluble free-radical initiators are preferably used. The kinetics and the mechanism are those for the corresponding bulk polymerization under similar conditions of temperature and initiator concentration.

Initiators affecting polymerization are those that are normally suitable for free-radical polymerization of acrylate monomers. Examples of such initiators include thermally-activated initiators such as azo compounds, hydroperoxides, peroxides and the like and photoinitiators such as benzophenone, benzoin ethyl ether and 2,2-dimethoxy-2-phenyl acetophenone. Other suitable initiators include lauroyl peroxide and bis(*t*-butyl cyclohexyl)peroxy dicarbonate. The initiator or mixture of initiators are present in a catalytically effective amount sufficient to bring about high monomer conversion in a predetermined time span and temperature range. Typically, the initiator is present in amounts ranging from 0.1 to approximately 2 parts per weight per 100 parts by weight of the polymerizable monomer starting materials.

Parameters that affect the concentration of initiator employed include the type of initiator and particular monomer and/or monomers involved. In addition mixtures of initiators may be used to control the rate of the polymerization reaction. It is believed that catalytically effective concentrations range from about 0.02 to about 2 percent by weight of the total monomers and more preferably, from about 0.20 to about 0.70 percent by weight monomers and/or monomers.

Optionally, a polymeric stabilizer may be used. Advantageously, the presence of the stabilizer permits the use of relatively low amounts of surfactant while still obtaining microspheres.

Any polymeric stabilizer that effectively provides sufficient stabilization of the final polymerized droplets and prevents agglomeration within a suspension polymerization

process is useful in the present invention. When used, a polymeric stabilizer will typically be present in the reaction mixture in an amount by weight of about 0.1 to about 3 parts by weight per 100 parts of polymerizable monomer, and more preferably will be present in an amount by weight of about 0.1 to about 1.5 parts by weight per 100 parts of polymerizable monomer.

Exemplary polymeric stabilizers include salts of polyacrylic acids of greater than 5000 molecular weight average (for example, ammonium, sodium, lithium and potassium salts), carboxy modified polyacrylamides (for example, Cyanamer™ A-370 from American Cyanamid), copolymers of acrylic acid and dimethylaminoethylmethacrylate and the like, polymeric quaternary amines (for example, General Aniline and Film's Gafquat™ 755, a quaternized polyvinylpyrrolidone copolymer, or Union Carbide's "JR-400", a quaternized amine substituted cellulosic), cellulose, carboxy-modified cellulose (for example, Hercules' Natrosol™ CMC Type 7L, sodium carboxy methylcellulose), and polyvinyl alcohols.

Surfactants will typically be present in the reaction mixture in an amount of no greater than about 5 parts by weight per 100 parts by weight of polymerizable monomer, preferably no greater than about 3 parts by weight, and most preferably in the range of 0.2 to 1.5 parts by weight per 100 parts by weight of polymerizable monomer.

Useful surfactants (also known as emulsifiers) include anionic, cationic, or nonionic surfactants and include but are not limited to anionic surfactants, such as alkylarylether sulfates and sulfonates such as sodium alkylarylether sulfate, e.g., Triton™ X200, available from Rohm and Haas, alkylarylpolyether sulfates and sulfonates such as alkylarylpoly(ethylene oxide) sulfates and sulfonates, preferably those having up to about 4 ethyleneoxy repeat units, and alkyl sulfates and sulfonates such as sodium lauryl sulfate, ammonium lauryl sulfate, triethanolamine lauryl sulfate, and sodium hexadecyl sulfate, alkyl ether sulfates and sulfonates such as ammonium lauryl ether sulfate, and alkylpolyether sulfate and sulfonates such as alkyl poly(ethylene oxide) sulfates and sulfonates, preferably those having up to about 4 ethyleneoxy units. Alkyl sulfates, alkyl ether sulfates, and alkylarylether sulfates are preferred. Additional anionic surfactants can include, for example, alkylaryl sulfates and sulfonates, for example sodium dodecylbenzene sulfate and sodium dodecylbenzene sulfonate, sodium and ammonium salts of alkyl sulfates, for example sodium lauryl sulfate, and ammonium lauryl sulfate; non-ionic surfactants, such as ethoxylated oleoyl alcohol and polyoxyethylene octylphenyl ether; and cationic surfactants, such as a mixture of alkyl dimethylbenzyl ammonium chlorides wherein the alkyl chain contains from 10 to 18 carbon atoms. Amphoteric surfactants are also useful in the present invention and include for example sulfobetaines, N-alkylaminopropionic acids, and N-alkylbetaines.

To initiate the polymerization reaction, a sufficient number of free radicals must be present. This may be achieved through several means that are well known in the art, such as heat or radiation free-radical initiation. For example, heat or radiation can be applied to initiate the polymerization of the monomers, which is an exothermic reaction. However, it is preferred to apply heat until thermal decomposition of the initiators generates a sufficient number of free radicals to begin the reaction. The temperature at which this occurs varies greatly depending upon the initiator used.

In addition, deoxygenation of the polymerization reaction mixture is often desirable. It is well known that oxygen dissolved in the reaction mixture can inhibit polymerization

and it is desirable to expel this dissolved oxygen. Although, an inert gas bubbled into the reaction vessel or through the reaction mixture is an effective means of deoxygenation, other techniques for deoxygenation that are compatible with suspension polymerization can be used. Typically, nitrogen is used to deoxygenate, although any of the Group VIIIA (CAS version) inert gases are also suitable.

While specific time and stirring speed parameters are dependent upon the types of monomers and initiators used, it is desirable to predisperse the reaction mixture until the reaction mixture reaches a state where the average monomer droplet size is between about 1 μm and 300 μm and preferably between 20 μm and 70 μm . The average particle size tends to decrease with increased and prolonged agitation of the reaction mixture.

Preferably, stirring and nitrogen purge are maintained throughout the reaction period. Initiation is begun by heating the reaction mixture. Following polymerization, the reaction mixture is cooled.

In a one-step process both the alkyl (meth)acrylate monomer and any optional comonomers are present together in the suspension at the initiation of polymerization. In a two-step process any optional comonomer is typically added after the initial exotherm resulting from polymerization of the alkyl (meth)acrylate monomer has peaked, but could be added at any point after polymerization has started. The other components, such as the initiator, stabilizers (if used), surfactants and modifiers are present in the reaction mixture as described in the processing steps herein above.

Following polymerization, a stable aqueous suspension of microspheres at room temperature is obtained. The suspension may have non-volatile solids contents of from about 10 to about 70 percent by weight. Upon prolonged standing, the suspension typically separates into two phases, one phase being an aqueous, essentially polymer microsphere-free phase and the other phase being an aqueous suspension of the polymeric microspheres, that is, the microsphere-rich phase. The aqueous suspension of microspheres may be utilized immediately following polymerization, because the suspension of microspheres of the present invention is particularly stable to agglomeration or coagulation. Advantageously, the microspheres of the present invention can be easily coated from an aqueous solution. Surprisingly, the microspheres of the present invention are well suited for conventional coating techniques and have enhanced fluid processing characteristics.

The microsphere-rich phase can be diluted with an additional amount of water or solvent, or redispersed upon shaking or other means of agitation. Generally, this aqueous suspension can be coated onto a backing or other substrate being employed using conventional coating methods, such as slot die coating, to provide an adhesive coating. The microspheres can be compounded with various rheology modifiers and/or latex adhesives or "binders". Typically, the adhesive coating which, when dried, exhibits a dry coating weight in the range of 0.2 to about 2 grams per square foot to provide an adhesive-coated sheet material in which the adhesive coating comprises polymeric microspheres, surfactant, and optionally polymeric stabilizers, rheology modifiers, and/or latex adhesives. Alternatively, the microspheres may be isolated and combined with an organic solvent if desired prior to coating them onto the backing.

Properties of the adhesive microspheres of the present invention can be altered by the addition of a tackifying resin(s) and/or plasticizer(s) after the polymerization. Preferred tackifiers and/or plasticizers for use herein include hydrogenated rosin esters commercially available from such

companies as Hercules, Inc. under the trade names of Foral™, Regalrez® and Pentalyn™. Tackifying resins also include those based on t-butyl styrene. Useful plasticizers include, but are not limited to: dioctyl phthalate, 2-ethylhexyl phosphate, tricresyl phosphate and the like. If such tackifiers and/or plasticizers are used, the amounts used in the adhesive mixture are amounts effective for the known uses of such additives.

Optionally, adjuvants, such as, rheology modifiers, colorants, fillers, stabilizers, pressure-sensitive latex binders and various other polymeric additives can be utilized. If such adjuvants are used, the amounts used in the adhesive mixture are amounts effective for the known uses of such adjuvants.

Backings used as substrates for adhesive articles may be materials that are conventionally used as a tape backing or may be of other flexible material. Such backings include, but are not limited to, those made from materials selected from the group consisting of poly(propylene), poly(ethylene), poly(vinyl chloride), polyester (e.g., poly(ethylene terephthalate), such as those available under the trade designation of "Scotch" film 8050 from 3M), polyamide films such as that available from DuPont Co., Wilmington, DE, under the trade designation "KAPTON," cellulose acetate, and ethyl cellulose. Backings may also be of woven fabric formed from threads of synthetic or natural materials such as cotton, nylon, rayon, glass, or ceramic material, or they may be of nonwoven fabric such as air laid webs of natural or synthetic fibers or blends of these. In addition, the backing may be formed of materials selected from the group consisting of metal, metallized polymeric film, and ceramic sheet material.

Preferred such materials include, but are not limited to, plastics such as polyethylene, polypropylene, polyesters, cellulose acetate, poly(vinyl chloride), and poly(vinylidene fluoride), as well as paper or other substrates coated or laminated with such plastics. These coated papers or thermoplastic films are often siliconized or otherwise treated to impart improved release characteristics. One or both sides of the backings or liners could have such release characteristics. Generally the backing or substrate material is about 50 µm to about 155 µm in thickness, although thicker and thinner backing or substrate materials are not precluded.

Particularly useful articles prepared using the adhesive microspheres of the present invention include repositionable adhesive products such as repositionable note and paper products, repositionable tape and tape flags, easel sheets, repositionable glue stick and the like, but may also include other non-repositionable industrial commercial, and medical adhesive products.

The present invention is further illustrated by the following examples, but the particular materials and amounts thereof recited in these examples, as well as other conditions and details, should not be construed to unduly limit this invention. All materials are commercially available or known to those skilled in the art unless otherwise stated or apparent. The following examples are illustrative in nature and are not intended to limit the invention in any way. All percentages listed in the Tables that follow are calculated on a weight basis to total monomer and vinyl unsaturated additive content.

EXAMPLES

Test Methods

Solvent Soluble Polymer Content

To determine the solvent soluble content of the prepared microspheres, the following process is used.

One gram of the water suspension of microspheres is dried in a vacuum oven without heat. After drying, 100 ml

of n-heptane is added and shaken for 24 hours. After shaking, the dispersion is poured through a filter paper (30 micrometer pores) to remove the non-soluble content. The filtrate is then dried in a 100° F. oven.

The weight of the dried filtrate divided by the dried suspension microspheres is the % solvent soluble polymer content. The test is repeated and the data is reported as the average of the number of trials.

Adhesive Microspheres Coating

A coating of the adhesive microspheres samples is made by allowing the suspension to cream. The cream (the microsphere-rich phase) is then coated through a 1 mil gap (25 micrometer) onto 62.5 micrometer matte polyester film (available from the 3M company as Scotch 8050).

Peel Adhesion to Bond Paper

Peel adhesion is the force required to remove a coated sheet from a bond paper substrate at a specific angle and rate of removal. In the examples this force is expressed in grams per one inch width of coated sheet. The procedure followed is:

A strip, one inch wide, of coated sheet is applied to the horizontal surface of 9.1 kilogram bond paper (21.8 cm. x 28.2 cm). A 2.04 kilogram hard rubber roller is used to firmly apply the strip to the bond paper. The free end of the coated sheet is attached to the adhesion tester load cell such that the angle of removal will be 90 degrees. The test plate is then clamped in the jaws of the tensile testing machine which is capable of moving the plate away from the load cell at a constant rate of 30.8 centimeters per minute. A load cell reading in grams per inch of coated sheet is recorded. The samples are tested three times. The average value of the three tests is reported.

GLOSSARY

- AA—acrylic acid
- AmA—ammonium acrylate
- ACM—acrylamide
- HEMA—hydroxyethyl methacrylate
- IOA—isooctyl acrylate
- IPA—isopropyl alcohol
- NVP—N-vinyl pyrrolidone
- RH—relative humidity
- NaSS—sodium styrene sulfonate
- AMPS—2-acrylamido-2-methylpropanesulfonate (ammonium salt)
- OAcM—N-tertiary octyl acrylamide
- DMAM—1,1-dimethyl-1(2-hydroxypropyl)amine methacrylimide
- PTSAN—para-toluene sulfonic acid (sodium salt)
- Lucido 75—benzoyl peroxyde with 25% water.

Example 1

To a two liter, three necked flask equipped with a thermometer mechanical stirrer and nitrogen inlet tube was charged 602.75 gm of deionized water, 35 gm of a 10% solids solution of Stepanol AMV (trade name for a 28% solids solution of ammonium lauryl sulfate commercially available from Stepan Company), 17.5 gm of a 10% solids solution of Goodrite K702 (trade name for a 25% solids solution of polyacrylic acid, 240,000 weight average molecular weight, commercially available from B.F. Goodrich Company) which had been neutralized to a pH of 7.0 with concentrated ammonium hydroxide, and 2.63 gm sodium styrene sulfonate. To this solution was added 347.37 gm of isooctyl acrylate and 1.05 gm of Perkadox 16N (trade name for a 95% active bis (4-tert-butylcyclohexyl) peroxydicarbonate initiator commercially available from AKZO

Chemicals Inc.) The agitation rate was set at 400 revolutions per minute (RPM), and the reaction mixture was heated to 50° C. and purged with nitrogen. The stirring, and nitrogen purge was maintained throughout the reaction period. The reaction was initially set at 50° C. exothermed after a few minutes and peaked at 77° C. The batch was maintained at 50° C. for 22 hours, cooled and filtered through a cheese cloth. The weight of the coagulum collected on the cheese cloth was measured and reported as % coagulum by weight to monomer and vinyl-unsaturated additive content. The particle size was 36 µm.

Example 2-8

The examples were prepared according to the procedure described in Example 1 except that the type and amount of emulsifier monomer, the amount of dodecanethiol and the amount and type of comonomer was changed. The formulations and test results are summarized in Tables 1 and 2 below.

TABLE 1

Example	Vinyl-unsaturated additive	Comonomer	% Dodecanethiol
1	0.75% NaSS	None	0
2	0.75% AMPS	None	0
3	1.5% AMPS	None	0
4	1% NaSS	2% NVP	0.04
5	0.75% NaSS	2% HEMA	0.02
6	1% NaSS	2% AcM	0.05
7	1% NaSS	12% OAcM	0
8	1% NaSS	12% OAcM	0.02

TABLE 2

Example	Adhesion to Bond Paper	% Coagulum	Particle Size (µm)	% Solvent Soluble Portion
1	199	0.02	36	7
2	NA	100	NA	NA
3	187	0.05	65	10
4	190	0.04	40	51
5	277	0.06	54	26
6	456	0.04	47	70
7	190	0.02	35	15
8	184	0.03	34	21

Comparative Examples C1-C9

These examples were prepared according to the procedure described in Example 1 except the vinyl-unsaturated additive was omitted. The formulations and test results are summarized in Tables 3 and 4 below.

TABLE 3

Example	Comonomer	% Dodecanethiol
C1	None	0
C2	0.75% DMAM	0
C3	0.75% AA	0
C4	0.75% AmA	0
C5	1.5% NVP	0.05
C6	1.5% HEMA	0.03
C7	1.5% AcM	0.05
C8	12% OAcM	0
C9	6% OAcM	0.02

TABLE 4

Example	Adhesion to Bond Paper	% Coagulum	Particle Size (µm)	% Solvent Soluble Portion
C1	NA	100	NA	NA
C2	159	0.17	~40*	15
C3	170	0.02	44	14
C4	205	0.03	70	16
C5	476	0.19	71	62
C6	417	0.19	62	60
C7	262	0.06	48	38
C8	NA	100	NA	NA
C9	NA	100	NA	NA

The results in Tables 1 and 2 show that the samples of this invention with a low level of vinyl-unsaturated additive result in microspheres which have enhanced adhesion to bond paper and are more stable during the polymerization exhibited by lower coagulum levels and are better repositionable adhesives.

The results with the comparative examples made without any vinyl unsaturated additive result in microspheres that entirely coagulate or, at best, have higher levels of coagulum and poorer adhesion to bond paper at comparable % solvent soluble levels.

The examples 1-8 and C1-C9 were prepared at 35% solids.

Comparative Examples C10

Example 1 from the EPO 0.209.337 B1 case was attempted as written in the case. However when 7.5 grams of 1-(3-sulphopropyl)-2-vinyl-pyridinium betaine (SPV) monomer was added to the cosolvent of 27.5 ml. of equal proportions of ethanol/toluene and agitated for two hours, the SPV did not dissolve. Since the example stated that dissolution would occur no further preparation of this example could be accomplished.

Example 9

To a two liter, three necked flask equipped with a thermometer, mechanical stirrer and nitrogen inlet tube was charged 507 gm of deionized water, 27 gm of a 10% solids solution of Stepanol AMV, 16 gm of a 10.7% solids solution of Goodrite K702 which had been neutralized to a pH of 7.0 with concentrated ammonium hydroxide, and 1.125 gm of sodium styrene sulfonate. To this solution was added 450 gm of isooctyl acrylate and 0.698 gm of Perkadox 16N and 0.698 gm of Lucidol 75. The reaction mixture was agitated at 400 RPM, heated to 45° C. and purged with nitrogen. The agitation and nitrogen purge was maintained throughout the reaction period. The reaction exothermed after a few minutes and peaked at 83° C. The batch was maintained at 60° C. for 4 hours, cooled and filtered through a cheese cloth. No coagulum was observed in the flask or on the cheese cloth. Particle size analysis indicated a mean volume diameter of 41 microns.

Examples 10 and 11

Examples 10, 11 and C11-C14 were repeated in a similar manner as that used in Example 9 except that the vinyl-unsaturated additive type and amount and the comonomer type and amount were varied as indicated in Table 5.

TABLE 5

Example	Vinyl-unsaturated additive	Comonomer	Stability
9	0.25% NaSS	None	stable
10	0.1% NaSS	None	agglomerated
C11	None	None	coagulated
C12	None	0.25% AmA	agglomerated
C13	0.25% PTSAN	None	agglomerated
C14	None	0.25% AA	~5% coagulum
11	0.1% NaSS	0.25% AA	stable

The results in Table 5 show that the sodium styrene sulfonate can stabilize an otherwise unstable one (compare Ex. 11 to C14 and Ex. 9 to C11, C12). Also the nonfunctionalized PTSAN does not exhibit the stabilizing effect that sodium styrene sulfonate does. All samples in Table 5 were made at 45% solids.

Example 12

To a two liter, three necked flask equipped with a thermometer, mechanical stirrer and nitrogen inlet tube was charged 259 gm of deionized water, 21 gm of a 10% solids solution of Stepanol AMV, 6 gm of a 10% solids solution of Goodrite K7200 (trade name for a 40% solids solution of poly(sodium acrylate), 20,000 weight average molecular weight, commercially available from B.F. Goodrich Company), and 3.5 gm of sodium styrene sulfonate. To this solution was added a solution of 329 gm of isooctyl acrylate, 5.25 g of HEMA, 5.25 g of NVP, 7 g of a 1% by weight solution of tert-dodecyl mercaptan dissolved in IOA, 0.123 gm of Perkadox 16N and 0.525 gm of Lucidol 75. The reaction mixture was agitated at 340 RPM, heated to 42° C. and purged with nitrogen. The agitation and nitrogen purge was maintained throughout the reaction period. The reaction exothermed after a few minutes and peaked at 75° C. The batch was maintained at 75° C. for 4 hours, cooled and filtered through a cheese cloth. No coagulum was observed in the flask or on the cheese cloth. Particle size analysis indicated a mean volume diameter of 45 microns. The % solvent soluble portion was 33%.

Example C15

Example C15 was carried out in a similar manner as Example 12 except that the NaSS was replaced with 3.5 g of AA which was subsequently neutralized with ammonium hydroxide to pH 7. Massive coagulation of the suspension occurred during the polymerization. Example C16 was carried out in a similar manner to that of Example 12 except that no NaSS was added. The reaction coagulated.

The results in Table 6 show that microspheres prepared without NaSS were not stable.

TABLE 6

Ex-ample	Vinyl-unsaturated additive	Co-monomer	Co-monomer	Co-monomer	Stability
12	1% NaSS	1.5% HEMA	1.5% NVP	None	stable
C15	None	1.5% HEMA	1.5% NVP	1% AmA	coagulated
C16	None	1.5% HEMA	1.5% NVP	None	agglomerated

The results in Table 6 show that without the use of sodium styrene sulfonate the suspension polymerization is not stable. The Ex. 12 and C15, 16 were made at 55% solids.

Example 13

Example 13 was prepared at 65% solids. This example illustrates that very high solids, low cost suspensions of adhesive microspheres can be made using the teachings of this invention.

To a two liter, three necked flask equipped with a thermometer, mechanical stirrer and nitrogen inlet tube was charged 160 gm of deionized water, 21 gm of a 10% solids solution of Stepanol AMV, 7.0 gm of a 10% solids solution of Goodrite K7200, and 1.75 gm of sodium styrene sulfonate. To this solution was added a solution of 346.5 gm of isooctyl acrylate, 1.75 g of AA, 0.070 gm of Perkadox 16N and 0.350 gm of Lucidol 75. The reaction mixture was agitated at 300 RPM, heated to 42° C. and purged with nitrogen. The agitation and nitrogen purge was maintained throughout the reaction period. The reaction exothermed after a few minutes an insulated jacket was then placed on the reaction flask. The reaction temperature peaked at 74° C. and the jacket was removed. The batch was maintained at 65° C. for 4 hours, cooled and filtered through a cheese cloth. The suspension filtered slowly (but completely) due to its relatively high viscosity. No coagulum was observed. Particle size analysis indicated a mean volume diameter of 57 micrometers. The % solvent soluble portion was 22%.

Various modifications and alterations of this invention will become apparent to those skilled in the art without departing from the scope and principles of this invention, and it should be understood that this invention should not be unduly limited to the illustrative embodiments set forth herein above. All publications and patents are incorporated herein by reference to the same extent as if each individual publication or patent was specifically and individually indicated to be incorporated by reference.

What is claimed:

1. A stabilized microsphere adhesive composition comprising:
 - (a) a plurality of polymeric, elastomeric microspheres wherein the microspheres are the reaction product of reactants comprising polymerizable starting materials comprising at least one C₄-C₁₄ alkyl (meth)acrylate monomer and optionally at least one comonomer,
 - (b) at least one vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms in an amount about 0.1 to 3 parts by weight of the microspheres;
 - (c) optionally, a polymeric stabilizer in an amount of between about 0.1 and about 3 parts by weight per 100 parts by weight of the microspheres;
 - (d) a surfactant in an amount of no greater than about 5 parts by weight per 100 parts by weight of the microspheres;
 - (e) optionally a modifier, wherein the modifier is at least one of a chain transfer agent, a tackifier, or a solvent in an amount that is sufficient to provide microspheres with a n-heptane soluble portion in the range of 30-98%; and
 - (f) an initiator in an amount effective to catalyze the polymerization reaction present in amounts ranging from about 0.1 to approximately 2 parts by weight per 100 parts by weight of the polymerizable monomer starting material.

2. The stabilized microsphere adhesive composition according to claim 1 wherein the vinyl-unsaturated additive is selected from the group consisting of the salts of sulfates of alpha-methylene carboxylic acids, the salts of sulfato-esters of alpha-methylene carboxylic acids, the salts of carboxy-terminated alkyl esters of alpha-methylene carboxylic acids, the salts of sulfoalkyl allyl ethers including 3-sulfo-2-hydroxy-1-propyl allyl ether, the salts of acrylamide alkane sulfonates, the salts of vinyl alkyl phosphonate esters, the salts of vinyl aryl sulfonates and mixtures thereof.

3. The stabilized microsphere adhesive according to claim 2 wherein the vinyl-unsaturated additive is a styrene sulfonate salt.

4. The stabilized microsphere adhesive according to claim 2 wherein the vinyl-unsaturated additive is a 2-acrylamido-2-methylpropane sulfonate salt.

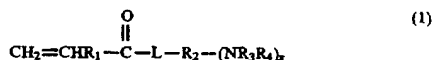
5. The stabilized microsphere adhesive composition according to claim 1 wherein the comonomer includes ionic, nonpolar monomers, polar monomers or mixtures thereof.

6. The stabilized microsphere adhesive composition according to claim 5 wherein the ionic monomers are selected from the group consisting of sodium methacrylate, ammonium acrylate, sodium acrylate, (I) trimethylamine p-vinyl benzimide, (II) 4,4, 9-trimethyl-4-azonia-7-oxo-8-oxa-dec-9-ene-1-sulphonate, (III) N,N-dimethyl-N-(beta-methacryloxyethyl) ammonium propionate betaine, (IV) trimethylamine methacrylimide, (V) 1,1-dimethyl-1-(2,3-dihydroxypropyl)amine methacrylimide; any zwitterionic monomer and mixtures thereof.

7. The stabilized microsphere adhesive composition according to claim 5 wherein the non-polar monomers are selected from the group consisting of 4-methyl-2-pentyl acrylate, 2-methylbutyl acrylate, isoamyl acrylate, sec-butyl acrylate, n-butyl acrylate, isodecyl methacrylate, t-butyl acrylate, t-butyl methacrylate, isobornyl acrylate, octyl acrylamide, methylmethacrylate, isononyl acrylate, isodecyl acrylate, styrene and the mixtures thereof.

8. The stabilized microsphere adhesive composition according to claim 5 wherein polar monomers may or may not contain a dissociable hydrogen and are selected from the group consisting of organic carboxylic acids comprising 3 to about 12 carbon atoms and having 1 to about 4 carboxylic acid moieties, acrylamide, methacrylamide, 2-hydroxyethyl acrylate, and mixtures thereof.

9. The stabilized microsphere adhesive composition according to claim 1 wherein the comonomer is an amino-functional monomer having the general formula:



wherein

R_1 is $-\text{H}$, $-\text{CH}_3$, $-\text{CH}_2\text{CH}_3$, cyano or carboxymethyl;

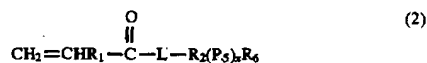
R_2 is a hydrocarbyl radical comprising 1 to about 12 carbon atoms;

R_3 and R_4 are independently H or an alkyl group containing 1 to about 12 carbon atoms or an arylalkyl group or together form a cyclic or heterocyclic moiety;

L is carbon-carbon bond, O, NH or S; and

x is an integer of 1 to 3.

10. The stabilized microsphere adhesive composition according to claim 1 wherein the comonomer is has the general formula:



wherein

R_1 is H, $-\text{CH}_3$, $-\text{CH}_2\text{CH}_3$, cyano or carboxymethyl;

R_2 is a hydrocarbyl radical comprising 1 to about 12 carbon atoms;

R_5 is $-\text{O}-$, alkylene oxide comprising 1 to 5 carbon atoms, or phenoxy oxide, wherein the alkylene oxide is $-\text{CH}_2\text{O}-$, $-\text{CH}_2\text{CH}_2\text{O}-$, or $-\text{CH}_2(\text{CH})\text{CH}_2\text{O}-$;

R_6 is H, $-\text{C}_6\text{H}_4\text{OH}$, or $-\text{CH}_3$

L is a carbon-carbon bond, O, NH or S; and

x is an integer with the proviso that when R_5 is $-\text{O}-$, x is an integer of 1-3.

11. The stabilized microsphere adhesive composition according to claim 1 wherein the comonomer is an amido-functional monomer general formula:



wherein

R_1 is H, $-\text{CH}_3$, $-\text{CH}_2\text{CH}_3$, cyano or carboxymethyl; and

R_3 , R_4 are independently H or an alkyl group containing 1 to about 12 carbons or an arylalkyl group or together form a cyclic or heterocyclic moiety.

12. The stabilized microsphere adhesive composition according to claim 1 wherein the comonomer is (meth)acrylonitrile, furfuryl (meth)acrylate and tetrahydrofurfuryl (meth)acrylate, 2-vinyl pyridine, or 4-vinyl pyridine.

13. The stabilized microsphere adhesive composition according to claim 1 wherein when a comonomer is present, the relative amounts by weight of the alkyl (meth)acrylate monomer(s) and the comonomer is in the range of about 99.5/0.5 to 75/25.

14. The stabilized microsphere adhesive composition according to claim 1 wherein the surfactant is in an amount of about 0.2 to 1.5 parts by weight per 100 parts by weight of the microspheres.

15. The stabilized microsphere adhesive composition according to claim 1 wherein the modifier is a chain transfer agent.

16. The stabilized microsphere adhesive composition according to claim 1 wherein the modifier is a solvent.

17. The stabilized microsphere adhesive composition according to claim 1 wherein the modifier is a tackifier.

18. The stabilized microsphere adhesive composition according to claim 1 wherein the modifier is a plasticizer.

19. A one step polymerization process for preparing polymeric elastomeric microspheres comprising the steps of:

(a) stirring or agitating a mixture comprising polymerizable monomer starting materials comprising:

(i) at least one C_4 - C_{14} alkyl (meth)acrylate monomer and, optionally at least one comonomer;

(ii) from 0.1 to 3 parts by weight of at least one vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms.

(iii) an initiator for the polymerizable monomer starting materials present in amounts ranging from 0.1 to approximately 2 parts per weight per 100 parts by weight of the polymerizable monomer starting materials;

(iv) optionally, a polymeric stabilizer in an amount in the range of 0.1 to about 3 parts by weight per 100

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parts by weight of the polymerizable monomer starting materials;

(v) a surfactant in an amount no greater than about 5 parts by weight per 100 parts by weight of polymerizable monomer;

(vi) water to form an oil in water suspension; and

(vii) optionally a modifier in an amount sufficient to provide a n-heptane soluble portion in the range of 30-98%; and

(b) polymerizing the (meth)acrylate monomer(s) and the comonomer(s), if present; wherein microspheres are provided.

20. A two step polymerization process for preparing polymeric elastomeric microspheres comprising the steps of:

(a) stirring or agitating a mixture comprising:

(i) at least one C_4 - C_{14} alkyl (meth)acrylate monomer;

(ii) from 0.1 to 3 parts by weight of at least one vinyl-unsaturated additive having both an ionic moiety and a hydrophobic moiety and containing at least 5 but not more than 40 carbon atoms.

(iii) an initiator for the monomer present in amounts ranging from 0.1 to approximately 2 parts per weight

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per 100 parts by weight of the polymerizable monomer starting materials;

(iv) optionally, a polymeric stabilizer in an amount in the range of 0.1 to about 3 parts by weight per 100 parts by weight of the polymerizable monomer starting materials;

(v) a surfactant in an amount of no greater than about 5 parts by weight per 100 parts by weight of the polymerizable monomer starting materials;

(vi) optionally a modifier in an amount sufficient to provide a n-heptane soluble portion in the range of 30-98%; and

(vii) water to form an oil in water suspension;

(b) at least partially polymerizing the polymerizable monomer starting materials;

(c) adding to the suspension at least one comonomer; and

(d) continuing the polymerization of the polymerizable monomer starting materials; wherein microspheres are provided.

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